

ENCRYPTED NANOPARTICLES FOR SECURITY, COUNTER-INDUSTRIAL
ESPIONAGE, AND COUNTERFEITING

A Dissertation

by

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ABSTRACT

Acts of terrorism and the use of explosives raise concerns about what can be done to prevent such acts. From stopping such a threat to preventing the illicit counterfeiting and smuggling of sensitive and proprietary goods, identification taggant technology can be applied to identify and interdict materials of a dubious nature. This cost effective approach to tag and track materials will find application in both the government and private sectors, particularly in the prevention of corporate espionage.

A taggant system was created based on the modification of the natural isotopic vector of cadmium and tellurium to create artificial, unique, and robust identification capabilities. A proof of concept trial was undertaken to ascertain the effectiveness, reliability, and integrity of a system. Stable isotopes were mixed in various combinations to alter the isotopic vector and then neutron activation analysis techniques were used to evaluate the quality of the taggants. Two different neutron activation analysis facilities of differing capabilities were employed to test the limitations of the method in detection of trace amounts of taggant material.

Testing of the taggant system found that solid phase taggants were capable of producing around 14,000 unique taggants for under \$250 each while liquid phase taggants were capable of producing around 2,000 unique taggants for under \$50 each. Common research reactors were also shown to be effective for use in a taggant system for certain isotopic vectors.

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Physics.

NOMENCLATURE

EOD	End of Decay
EOI	End of Irradiation
HFIR	High Flux Isotope Reactor
HPGe	High Purity Germanium
MS	Mass Spectrometry
NAA	Neutron Activation Analysis
NAAL	Neutron Activation Analysis Laboratory
ORNL	Oak Ridge National Laboratory
ppb	Part per Billion
ppm	Part per Million
RCT	Radiological Control Technician
TRIGA	Training, Research, Isotopes, General Atomic
VFX	Vertical Experiment Facility

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1. INTRODUCTION

1.1 Gum in the Smithsonian

At 8:01 AM on June 26, 1974, Clyde Dawson purchased a 10-pack of Wrigley's Juicy Fruit gum [1] from the Marsh supermarket in Troy, Ohio. It just happened to be the first item placed on the counter that day and scanned by cashier Sharon Buchanan. That pack of gum is now on display at the Smithsonian Institution's National Museum of American History as the first item scanned with the ubiquitous UPC barcode that is heavily used in supermarkets today. Barcodes changed the paradigm of purchasing products. Instead of manually reading the price from a sticker, the barcode was automatically read by a scanner and then associated with a price from a database. Inventory of items became routine and automated, allowing less burden on workers, while decreasing theft.

1.2 Oklahoma City

Besides streamlining the process of purchasing groceries, ways to tag and identify objects have aided in the investigation of serious capital crimes. The Alfred P. Murrah federal building was destroyed on April 19, 1995 using a Ryder moving truck filled with explosives [2]. The culprit, Timothy McVeigh, was pulled over only 90 minutes after the bombing occurred but not because of any connection with the bombing. He was missing a license plate and it was only after the mangled truck axle was found in the debris the next day was he connected to the crime. The VIN number from the axle was traced to the rental agency, which then implicated Mr. McVeigh in the bombing. The axle served as a key piece of evidence at his trial.

Years earlier, in 1979, a man named Nathan Allen was killed by a bomb made of dynamite [3] placed in his truck. Small microscopic particles called taggants placed in

the explosives were able to identify the batch and date of manufacture. Investigators eventually traced the sale of explosives to James McFillin, Nathan Allen's uncle, who was convicted of the bombing. The taggants used in the bombing were small flakes with layers of color. After an explosion, these particles were inspected under a microscope and the sequence of the layers of colors were used to identify which batch the explosives came from and sales records were used to identify James McFillin as the purchaser.

If Timothy McVeigh had chosen, he could have located the VIN number on the axle and removed it before the bombing. This would be an arduous task, but still possible as he could locate the VIN number through simple inspection of the axle. He may have been convicted through other means or evidence, but one less link to the crime would have bolstered his case. Mr. McFillin would have a more difficult time locating each microscopic taggant placed in the dynamite, aside from the fact that he was most likely unaware of their presence when purchasing the dynamite. Small taggants that are not readily observable serve a purpose to solve crimes and track objects without the possibility of unwanted detection. Technology has evolved to the point that simple sticker barcodes are no longer adequate to protect against counterfeiting and malevolent acts. A more complex system is needed to help prevent forgery and also to help investigators solve crimes.

1.3 Isotopic Taggants

Isotopic ratios and vectors in samples have long been used in forensic applications [4, 5, 6]. An isotopic vector is the mathematical vector of values representing the atomic fraction of a set of isotopes present in an element. Just about everything from common packaging tape [7] to different brands of Mexican cigarettes [8] to firearm residue [9] have been forensically studied through the use of isotopic ratios, usually

to determine the location of origin or manufacture of the material. While mass spectrometry (MS) represents the pinnacle of sensitivity for forensic applications [5], neutron activation analysis (NAA) also has value as a tool in forensic studies.

Recent ideas have called for the use of ratios to designate a unique identifier to an object or product in either chemical [10] or isotopic [11] form, but development of such a system lacks significant study in systems dealing with few elements, increasing the correlation between isotopic ratios within an element. While the error of the analytical techniques are well known and understood [12, 13, 14], the error associated with such a taggant system is not yet understood, especially for few element systems.

To aid in the tracking and recovery of illicit materials, a system to create a discreet marking, or taggant, to identify objects is desirable. Such a taggant system would mimic the form and function of traditional paper taggants used presently, with the minor difference that the taggants be unobtrusive and almost invisible to any common onlooker. Paper taggants would be much too large and susceptible to tampering. With the ability to adjust the isotopic composition of just about every element, the composition of an element in or on the material could be used as a unique identifier. Through the perturbation of the isotopic enrichment of just a few elements on the periodic table, a database of possible combinations of isotopes could be created and the compositions of these isotopes would determine a unique taggant. The original concept was put forward by Mirzadeh, Garland, and Rondinone in a patent disclosure which was filed with the ORNL Patent Office in January of 2009. Changing the taggant at the atomic level would allow the system to have a higher degree of integrity compared to traditional paper taggants and tags. Taggants can be read and deciphered through scientific analysis of the isotopic composition. The material would be placed on or about the object in a location known only to the manufacturer, ensuring that the integrity remains intact.

A proof of concept study was conducted to assess a potential taggant system. Elements were chosen and the natural isotopic abundance was perturbed to produce different isotopic vectors and taggants. These taggants were then read using NAA to evaluate if such a system utilizing isotopic perturbation is feasible and also to quantify the error arising from the system.

2. NEUTRON ACTIVATION ANALYSIS

2.1 History of Neutron Activation Analysis

2.1.1 The Hidden Nobel Prizes

Max Von Laue and James Franck were German physicists that were awarded the Nobel Prize in 1914 and 1925, respectively, for their work in physics. When war broke out, they sent their prizes for safekeeping to a Hungarian radiochemist, George de Hevesy [15], who was practicing at the Neils Bohr Institute in Copenhagen, Denmark. After Germany invaded Denmark, it became illegal to send gold out of the country. Not wanting the gold to become Nazi property, Dr. de Hevesy dissolved the prizes in a solution of aqua regia and left the solution on a shelf at the institute until the war ended. Upon returning to the institute, Dr. de Hevesy precipitated the gold and the prizes were recast by the Nobel Society.

2.1.2 Rutherford and Radium D

Long before he was hiding Nobel Prizes in plain sight, de Hevesy was attempting to separate lead from lead. While working for Ernest Rutherford, de Hevesy was asked to perform a task. Rutherford had acquired a quantity of Radium D, a byproduct of radium. To make use of this Radium D, de Hevesy would need to extract it from the large quantity of lead that it was in. After a year of failure, de Hevesy gave up considering the experiment a failure and regretting his young optimism that he had when he had accepted the assignment from Rutherford.

To make the most of a bad situation, de Hevesy thought instead to label the large quantity of lead with even more Radium D and he spiked the lead with Radium D that he had obtained from radon. He was able to label lead through the first use

of a radiotracer method. This radiotracer method was the first step that led to the proposal of NAA by de Hevesy and Levi [16] in 1936. Seaborg extended the idea to charged particle activation analysis in 1938, but the method of NAA remained stagnant until the development of nuclear reactors in the 1940s that could produce a neutron fluence rate on the order of 1×10^{12} neutrons $\text{cm}^{-2} \text{s}^{-1}$. With the advent of research reactors, NAA became popular in the 1960s as an analytical technique offering parts per million (ppm) and parts per billion (ppb) detection limits for most elements of the periodic system. It is even used to this day to detect ^{210}Pb , a radioactive isotope of lead, which was incorrectly known during de Hevesy's time as Radium D.

2.2 Activation Analysis

The method of NAA is one in which a stable isotope is subjected to a strong neutron field, usually in a nuclear reactor, and transmuted into a radioactive isotope that decays by emitting one or more characteristic gamma rays that can identify the material [16]. Most radioactive isotopes are formed through neutron capture reactions in which a stable nuclide absorbs a neutron and forms a heavier radioactive nuclide, most usually releasing a gamma ray in the process. Other less common reactions may also take place, resulting in the release of an alpha particle, proton, or light nuclide.

NAA has the ability to distinguish individual isotopes of a sample without the need to chemically extract or process the sample. This is advantageous in the analysis of a sample with a complex chemical matrix or for samples that need to be preserved. Historical artifacts have been studied with only light exposure to a neutron field, allowing nondestructive analysis of these items.

MS has caught up with and exceeded NAA in terms of precision and accuracy

for trace elemental analysis. While MS has a lower detection limit [17], NAA has the advantage of being faster and cheaper than MS [5] due to the shorter sample preparation times needed for NAA and the ability to irradiate samples with little or no chemical processing during preparation. NAA is free of isobaric interference because nuclides that are similar in mass will not necessarily have characteristic gamma rays that will interfere with each other. NAA is frequently used for analysis of elements with similar mass, providing an easy way to obtain an isotopic vector of an element.

2.3 Activation Equations

2.3.1 Bateman-Rubinson Equations

Harry Bateman first solved a system of differential equations [18] to model the radioactive buildup and decay of naturally radioactive elements. The neutron wasn't discovered until 1932. It was only after this discovery that William Robinson at Brookhaven National Laboratory incorporated neutron activation into the Bateman equations to form the Bateman-Rubinson equations [19, 20, 21] to model the buildup and decay of radionuclides in a neutron field. The general solution to the Bateman-Rubinson equations are given by:

$$N_n(t_b) = \Lambda_1^* \Lambda_2^* \dots \Lambda_{n-1}^* N_1^0 \sum_{i=1}^{i=n} C_i \exp(-\Lambda_i t_b) \quad (2.1)$$

where

$$C_i = \prod_{j=1}^{j=i} \frac{1}{\Lambda_j - \Lambda_i} (j \neq i) \quad (2.2)$$

where

t_b is the length of the irradiation time;

$N_n(t_b)$ represents the number of atoms of nuclide n present at time t_b ;
 Λ_n^* is the modified disappearance constant, $\lambda + \phi\sigma$, representing removal either by radioactive decay, nuclear reaction, or a combination of both;
 λ is the decay constant or partial decay constant in case of branching decay;
 ϕ is the neutron fluence rate; and
 σ is the reaction cross section or partial reaction cross section in case of branching activation.

For purposes of NAA, a simple case of transformation in a neutron field can be applied where a stable nuclide is transformed into a radioactive nuclide that subsequently decays to a different stable nuclide. If $\lambda_2 \gg \phi\sigma_1$, which applies to almost all cases of NAA where the neutron fluence and cross section are not extremely high with an extremely long irradiation time, then Eq. 2.1 and 2.2 simplify to

$$N_2(t_b) = \frac{\phi\sigma_1 N_1^0}{\lambda_2} [1 - \exp(-\lambda_2 t_b)]. \quad (2.3)$$

Rearrangement of Eq. 2.3, with an expansion to two-group theory yields:

$$N_1^0 = \frac{A_2(t_b)}{(\sigma_{1,t}\phi_t + \sigma_{1,r}\phi_r)[1 - \exp(-\lambda_2 t_b)]} \quad (2.4)$$

where

N_1^0 is the number of atoms of the initial nuclide before irradiation;

t_b is the length of time the sample was irradiated;

$A_2(t_b)$ is the activity of the resulting radionuclide at time t_b ;

$\sigma_{1,t}$ is the thermal neutron cross section of the initial nuclide;

ϕ_t is the thermal energy neutron fluence rate;

$\sigma_{1,r}$ is the resonance energy neutron cross section of the initial nuclide;

ϕ_r is the resonance energy neutron fluence rate; and
 λ_2 is the decay constant of the radionuclide.

Eq. 2.4 is widely used in NAA for simple activation of a stable nuclide in a neutron field for a relatively short amount of time. This model assumes that either the neutron field is not strong enough or the sample is not irradiated for such a period of time as to destroy a significant amount of the initial sample. Corrections for target burnup [20, 21] may be used for samples that experience a significant amount of irradiation. The model also assumes that the neutron fluence rate remains constant throughout irradiation.

To determine the variance associated with the irradiation of a sample, the general formula for error propagation [22] for a general function $X = f(A, B, C, \dots)$ is given by:

$$\sigma_X^2 = \left(\frac{\partial f}{\partial A} \sigma_A \right)^2 + \left(\frac{\partial f}{\partial B} \sigma_B \right)^2 + \left(\frac{\partial f}{\partial C} \sigma_C \right)^2 + \dots \quad (2.5)$$

where

σ_X^2 is the variance of X . Eq. 2.5 assumes that the errors have a symmetric Gaussian distribution around the mean and that the variables are independent [23]. Using Eq. 2.5, the variance associated with irradiation of a sample using Eq. 2.4 is given by:

$$\begin{aligned}
S_N^2 = & \frac{S_A^2}{(\sigma_t\phi_t + \sigma_r\phi_r)^2(1 - \exp(-\lambda t))^2} + \frac{S_{\phi_t}^2\sigma_t^2A^2}{(\sigma_t\phi_t + \sigma_r\phi_r)^4(1 - \exp(-\lambda t))^2} \\
& + \frac{S_{\phi_r}^2\sigma_r^2A^2}{(\sigma_t\phi_t + \sigma_r\phi_r)^4(1 - \exp(-\lambda t))^2} + \frac{S_{\sigma_t}^2\phi_t^2A^2}{(\sigma_t\phi_t + \sigma_r\phi_r)^4(1 - \exp(-\lambda t))^2} \\
& + \frac{S_{\sigma_r}^2\phi_r^2A^2}{(\sigma_t\phi_t + \sigma_r\phi_r)^4(1 - \exp(-\lambda t))^2} + \frac{S_t^2A^2\lambda^2\exp(-2\lambda t)}{(\sigma_t\phi_t + \sigma_r\phi_r)^2(1 - \exp(-\lambda t))^4} \\
& + \frac{S_\lambda^2t^2A^2\exp(-2\lambda t)}{(\sigma_t\phi_t + \sigma_r\phi_r)^2(1 - \exp(-\lambda t))^4} \quad (2.6)
\end{aligned}$$

where

S is the standard deviation.

2.3.2 Parametric and Comparative NAA

In the method of parametric NAA, a sample is irradiated in a known neutron field and counted on a characterized and calibrated detector. Using Eq. 2.4, the isotopic vector of the sample is calculated. The inherent characteristic of this method is that all nuclear properties of the involved nuclides must be accurately known, specifically the cross section, neutron fluence rate, and decay constant. Additionally, the detector system used to determine the activity of the resulting radionuclide must be well characterized and calibrated using check sources such as a multi-isotope europium source.

In the method of comparative NAA, a known reference sample is irradiated and counted. The unknown sample is then irradiated and counted in the same manner as the known reference sample. The resulting gamma spectra obtained from both samples are compared. By taking ratios of the standard and the unknown, many of the variables in Eq. 2.4 cancel out, resulting in

$$N_i = \frac{A_i}{A_{\text{std}}} \frac{m_{\text{std}} N_A}{M_{\text{mi}}} \quad (2.7)$$

where

N_i is the number of atoms of the unknown nuclide i ;

A_i is the decay corrected activity at end of irradiation of the unknown nuclide i ;

A_{std} is the decay corrected activity at end of irradiation of the known nuclide standard;

m_{std} is the mass of the radioisotope precursor in the standard;

N_A is Avogadro's number, $6.02214179(30) \times 10^{23}$ [24]; and

M_{mi} is the atomic weight of the nuclide i .

Using Eq. 2.5, the associated standard deviation of N_i in Eq. 2.7 is expressed by [25]:

$$S_{N_i} = N_i \sqrt{\left(\frac{S_{A_i}}{A_i}\right)^2 + \left(\frac{S_{A_{\text{std}}}}{A_{\text{std}}}\right)^2 + \left(\frac{S_{m_{\text{std}}}}{m_{\text{std}}}\right)^2 + \left(\frac{S_{N_A}}{N_A}\right)^2 + \left(\frac{S_{M_{\text{mi}}}}{M_{\text{mi}}}\right)^2}. \quad (2.8)$$

2.4 Advantage of Comparative NAA

Comparative NAA is superior to parametric NAA in many situations if a reference standard is available because the errors are reduced through the elimination of many variables. This method does require prior knowledge of the sample composition to obtain an accurate reference standard. For situations involving complicated buildup and decay schemes, comparative NAA is most useful as each sample will undergo the same complicated transmutations. For thick sample irradiation containers that will perturb the neutron fluence rate or energy, comparative NAA eliminates the

need to measure the neutron spectra for different sample containers. As long as the samples are irradiated and counted in the same conditions, such as irradiation time, detector geometry, similar sample containers, and neutron fluence rate, the comparative NAA method will result in less error than parametric NAA. This is based upon the assumption that the unknown sample is in a similar chemical matrix as the standard and that they are roughly similar in mass or thickness to prevent a significant difference in self-shielding in either the unknown or standard.

3. SELECTION OF NUCLIDES

3.1 Properties of Desirable Elements

To choose suitable elements for the formation of a taggant, a list of desired properties of possible elements was created:

- An element should be polyisotopic, with multiple activation products. The larger number of isotopes allows a greater number of possible combinations.
- Stable isotopes should generally not be contiguous as this eliminates many (n,γ) reactions from producing radionuclides. While other reactions such as (n,α) and (n,p) may produce radionuclides, those reactions are threshold reactions that require incident neutrons of higher energy and are generally less efficient at activation and transmutation in a thermal neutron environment due to their much smaller cross sections. While titanium has 5 stable isotopes, it is not a good candidate because all of them are located contiguously between ^{46}Ti and ^{50}Ti [26].
- Each activation product should originate from one stable parent. This allows effective attribution of each radionuclide to a parent for taggant decryption. An example of this would be ^{56}Mn produced by both $^{59}\text{Co}(n,\alpha)^{56}\text{Mn}$ and $^{55}\text{Mn}(n,\gamma)^{56}\text{Mn}$.
- The half-life of any resulting activation product should be conducive to NAA. A long half-life is usually associated with lower specific activity and will be harder to accurately detect in a short amount of time. A short half-life will allow the radionuclide to completely decay away before counting can begin. A half-life of a few hours to a few days is desired to allow both a moderate and

detectable specific activity and to also allow enough time for sample counting preparation.

- The cross sections of the nuclides should be roughly similar so that the ratio between two isotopes will not be significantly large. A strong difference in cross sections will allow some radionuclides to dominate with a high specific activity, which may hinder the accurate counting of other radionuclides.
- Elements should be chosen in such a way that they can form chemically stable compounds with each other. As taggants will consist of combinations of isotopes within combinations of elements, fractionation, or separation, of the elements and their respective isotopes through chemical processes will skew the accuracy of the taggant.

Economic factors must be considered in addition to the scientific traits that are desirable of chosen elements. The elements and isotopes must not be so expensive that they would not be of practical use for their intended purpose.

3.2 Choice of Cadmium and Tellurium Elements

A combination of nuclides drawn from the elements of cadmium (Cd) and tellurium (Te) were chosen as the materials of choice for the taggant system. A list of isotopes of interest [27] are shown in Table 3.1 with their natural abundance and in Table 3.2 with their atomic mass [28]. The standard atomic weight [24] of cadmium is $112.411 \pm 0.008 \text{ g mol}^{-1}$ and the standard atomic weight of tellurium is $127.60 \pm 0.03 \text{ g mol}^{-1}$.

Possible activation products and associated nuclear properties of these products [27, 29] are shown in Table 3.3. Some nuclides lead to only one activation product while others lead to two activation products, but no two stable nuclides produce the

Table 3.1: Natural abundance [27] of selected stable isotopes of Cd and Te

Isotope	Natural Abundance (%)
¹⁰⁶ Cd	1.25 ± 0.04
¹⁰⁸ Cd	0.89 ± 0.02
¹¹² Cd	24.13 ± 0.14
¹¹⁴ Cd	28.73 ± 0.28
¹¹⁶ Cd	7.49 ± 0.12
¹²⁰ Te	0.096 ± 0.002
¹²² Te	2.603 ± 0.004
¹²⁴ Te	4.816 ± 0.006
¹²⁶ Te	18.95 ± 0.01
¹²⁸ Te	31.69 ± 0.01
¹³⁰ Te	33.8 ± 0.01

Table 3.2: Atomic mass of selected stable isotopes [28] of Cd and Te

Isotope	Atomic Mass (u±μu)
¹⁰⁶ Cd	105.906459 ± 6
¹⁰⁸ Cd	107.904184 ± 6
¹¹² Cd	111.9027578 ± 2.9
¹¹⁴ Cd	113.9033585 ± 2.9
¹¹⁶ Cd	115.904756 ± 3
¹²⁰ Te	119.904020 ± 10
¹²² Te	121.9030439 ± 1.6
¹²⁴ Te	123.9028179 ± 1.6
¹²⁶ Te	125.9033117 ± 1.6
¹²⁸ Te	127.9044631 ± 1.9
¹³⁰ Te	129.9062244 ± 2.1

same activation product, allowing simple attribution to one stable nuclide.

The half-lives listed in Table 3.3 are conducive to NAA. The cross sections are not significantly different such that one isotope would dominate the NAA process.

Table 3.3: Activation products of Cd and Te

Target Isotope	Activation Product	Half-Life of the Activation Product	Thermal Energy Cross Section (b)	Resonance Energy Cross Section (b)
^{106}Cd	^{107}Cd	6.5 ± 0.02 h	1	4.1
^{108}Cd	^{109}Cd	462.6 ± 0.4 d	0.72 ± 0.13	10.7
^{112}Cd	$^{113\text{m}}\text{Cd}$	14.1 ± 0.5 y	2.2 ± 0.5	12.5 ± 1
^{114}Cd	^{115}Cd	53.46 ± 0.1 h	0.294 ± 0.016	12.6 ± 1
^{114}Cd	$^{115\text{m}}\text{Cd}$	44.6 ± 0.3 d	0.036 ± 0.007	12.6 ± 1
^{116}Cd	^{117}Cd	2.49 ± 0.04 h	0.05 ± 0.008	1.5 ± 0.02
^{116}Cd	$^{117\text{m}}\text{Cd}$	3.36 ± 0.05 h	0.025 ± 0.01	1.5 ± 0.02
^{120}Te	^{121}Te	16.78 ± 0.35 d	2 ± 0.3	0
^{120}Te	$^{121\text{m}}\text{Te}$	154 ± 7 d	0.34 ± 0.06	0
^{122}Te	$^{123\text{m}}\text{Te}$	119.7 ± 0.1 d	0.49 ± 0.08	88 ± 10
^{124}Te	$^{125\text{m}}\text{Te}$	57.4 ± 0.15 d	0.04 ± 0.025	5.3 ± 0.7
^{126}Te	^{127}Te	9.35 ± 0.07 h	0.305 ± 0.064	8 ± 0.6
^{126}Te	$^{127\text{m}}\text{Te}$	109 ± 2 d	0.135 ± 0.023	8 ± 0.6
^{128}Te	^{129}Te	69.6 ± 0.3 m	0.18 ± 0.008	1.58 ± 0.06
^{128}Te	$^{129\text{m}}\text{Te}$	33.6 ± 0.1 d	0.02 ± 0.002	77 ± 5
^{130}Te	^{131}Te	25 ± 0.1 m	0.185 ± 0.01	0.4 ± 0.05
^{130}Te	$^{131\text{m}}\text{Te}$	30 ± 2 h	0.01 ± 0.003	0.4 ± 0.05

3.2.1 Nanoparticles

3.2.1.1 Applications of Nanoparticles

Nanoparticles are finding new uses for many applications. Everything from printer ink to cancer therapy is made possible through the use of nanoparticles. Experience with the manufacture of CdTe nanoparticles [30] has shown to be effective for ease of application to just about any surface. For this reason, nanoparticles were chosen as the medium of a taggant system to discreetly mark an object. Nanoparticles are in a stable chemical configuration to prevent the fractionation of elements, which would alter or skew the accuracy of the taggant. When the item is recovered,

the nanoparticles would be removed and analyzed to determine the origin of the item. Only a portion of the particles would need to be recovered from the item, allowing for multiple readings with no damage to the item.

3.2.1.2 Experimentation without Nanoparticles

Although nanoparticles are the intended final form of a taggant system, they were not used during experimentation. The additional processing required to produce nanoparticles was not needed because NAA is impervious to the chemical and structural form of the material. This is also a benefit, allowing chemical and material flexibility of the system.

3.3 Production of Enriched Stable Isotopes

3.3.1 Manhattan Project

During the second World War, an electromagnetic process [31] was employed at the Y-12 plant in Oak Ridge, TN to separate isotopes of uranium using Calutrons. In this process, uranium is first ionized by bombarding it with electrons, and the beam of ionized uranium is then passed through a large magnetic field. The resulting isotopes are collected on plates for later processing. This method of uranium enrichment was replaced after the war with more efficient methods such as gaseous diffusion and gas centrifuges that could produce a higher volume of enriched uranium.

3.3.2 After the War

After the task of enriching uranium was moved elsewhere, the Calutrons became obsolete for the production of weapons and most were eventually shut down and removed from their buildings. The Calutrons in the Beta 3 building were repurposed for the experimental separation of other isotopes. Over the last half of the twentieth century, many isotopes were separated and made available to the general public for

scientific research purposes, including many isotopes of cadmium and tellurium.

3.4 Procurement of Enriched Stable Isotopes

3.4.1 Definitions

To fully understand the process of purchasing enriched stable isotopes, a few things are defined for clarity.

- Atomic Mass - The mass of an individual nuclide. This is a fundamental property of the nuclide and will not change with enrichment.
- Atomic Weight (Relative Atomic Mass) - The weighted average of individual nuclide atomic masses that constitute an element. This value will change after a natural element is enriched.
- Standard Atomic Weight - A value of atomic weight for a given natural element that is representative of the element as found on earth. While the atomic weight of an element may vary across different locations on earth, the standard atomic weight is a representative value of all locations on earth.
- Isotopic Vector - A mathematical vector of values representing the atomic fraction of a set of isotopes present in an element.
- Batch - A sample of an enriched isotope containing a particular isotopic vector. Batches are identified by their assigned batch number or the primary isotope of enrichment.
- Sample - A prepared mixture of batches to form a taggant with a characteristic isotopic vector and taggant vector.
- Gravimetric Factor - The fractional presence of an element in a compound.

3.4.2 Impure Isotopes

Table 3.4: Enriched batches of cadmium and their isotopic vector (%)

Batch	¹⁰⁶ Cd	¹⁰⁸ Cd	¹¹⁰ Cd	¹¹¹ Cd	¹¹² Cd	¹¹³ Cd	¹¹⁴ Cd	¹¹⁶ Cd
Natural	1.25	0.89	12.49	12.80	24.13	12.22	28.73	7.49
155201	88.4	0.48	2.02	1.61	2.49	1.14	2.50	1.36
213001	79.01	0.68	3.03	2.60	5.56	1.73	6.21	1.18
lx1440b	0.70	67.3	13.8	4.7	5.7	2.35	4.4	1.12
222101	0.01	0.01	0.086	0.183	0.37	0.48	98.69	0.178
190842	0.01	0.01	0.08	0.19	0.40	0.60	98.55	0.19

The electromagnetic enrichment process is not perfectly efficient because of the limited number of stages of Calutrons used for enrichment. More abundant isotopes achieve a higher final enrichment faster [31] than isotopes that constitute a minute natural fraction of an element. Isotopes with small abundances will also tend to cost more [32] because they need more feedstock to produce a given amount of product. When purchasing a stable isotope, all other naturally occurring isotopes of the element are usually still present, but in smaller quantities. This is not always true, as isotopes close in mass to the primary enriched isotope may also be enriched slightly instead of depleted. A list of batches of cadmium and their enrichment are shown in Table 3.4 with the uncertainty shown in Table 3.5.

Similar information concerning tellurium is found in Tables 3.6 and 3.7.

MS is performed to determine both the isotopic vector and associated uncertainty of a batch. The MS results are included with the purchase of a batch. The primary isotope of each batch is shown in Table 3.8. The primary isotope is prominent in tellurium batches, comprising over 90% of the batch. The primary isotope is less prominent among some cadmium batches, particularly batch lx1440b.

Table 3.5: Enriched batches of cadmium and their isotopic vector uncertainty (%)

Batch	^{106}Cd	^{108}Cd	^{110}Cd	^{111}Cd	^{112}Cd	^{113}Cd	^{114}Cd	^{116}Cd
155201	0.1	0.05	0.05	0.05	0.05	0.05	0.05	0.05
213001	0.05	0.01	0.02	0.01	0.01	0.01	0.01	0.01
lx1440b	0.05	0.1	0.1	0.1	0.1	0.05	0.1	0.05
222101	0	0	0.003	0.005	0.01	0.01	0.02	0.005
190842	0	0	0.01	0.01	0.01	0.02	0.05	0.01

Table 3.6: Enriched batches of tellurium and their isotopic vector (%)

Batch	^{120}Te	^{122}Te	^{123}Te	^{124}Te	^{125}Te	^{126}Te	^{128}Te	^{130}Te
Natural	0.09	2.55	0.89	4.74	7.07	18.84	31.74	34.08
201801	0.05	96.45	0.14	0.34	0.45	0.88	0.97	0.77
206246	0.06	94.23	0.32	0.92	0.50	1.55	1.26	1.16
209026	0.13	0.11	0.07	94.81	0.93	2.55	0.86	0.55
ca297a	0	0.1	0	0.6	0.7	95.4	2.6	0.6
187146	0.02	0.03	0.03	0.2	0.24	98.02	0.94	0.52
166401	0.04	0.01	0.02	0.03	0.04	0.17	99.19	0.54
192241	0.01	0.01	0.01	0.03	0.05	0.20	98.77	0.92
187340	0.01	0.02	0.01	0.03	0.06	0.21	1.23	98.43

Table 3.7: Enriched batches of tellurium and their isotopic vector uncertainty (%)

Batch	^{120}Te	^{122}Te	^{123}Te	^{124}Te	^{125}Te	^{126}Te	^{128}Te	^{130}Te
201801	0	0.1	0.01	0.02	0.02	0.05	0.05	0.05
206246	0.01	0.13	0.05	0.02	0.01	0.04	0.01	0.01
209026	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
ca297a	0	0.1	0	0.2	0.2	0.8	0.5	0.2
187146	0.01	0.01	0.02	0.01	0.01	0.11	0.07	0.05
166401	0	0.01	0.01	0.01	0.01	0.01	0.06	0.02
192241	0.01	0.01	0.01	0.01	0.01	0.01	0.03	0.03
187340	0.01	0.01	0.01	0.01	0.01	0.01	0.03	0.03

Table 3.8: Primary isotope of enriched batches

Batch	Primary Isotope
155201	^{106}Cd
213001	^{106}Cd
lx1440b	^{108}Cd
222101	^{114}Cd
190842	^{114}Cd
201801	^{122}Te
206246	^{122}Te
209026	^{124}Te
ca297a	^{126}Te
187146	^{126}Te
166401	^{128}Te
192241	^{128}Te
187340	^{130}Te

3.4.3 Final Selection of Isotopes

Table 3.9: Price of stable isotope batches

Batch	Price ($\text{\$ mg}^{-1}$)
^{106}Cd	76.68
^{108}Cd	100.81
^{112}Cd	2.98
^{114}Cd	2.09
^{116}Cd	19.02
^{120}Te	1,741.45
^{122}Te	78.92
^{124}Te	31.94
^{126}Te	9.58
^{128}Te	4.71
^{130}Te	4.81

A factor in the choice of isotopes for use in a taggant system was the price and

availability of each batch. The price of selected batches are shown in Table 3.9. The primary isotope is shown instead of the batch number.

From examination of Table 3.9, the decision was made to exclude ^{120}Te from a possible taggant system. The price was prohibitive due to the low natural abundance of ^{120}Te as shown in Table 3.1, making ^{120}Te one of the most expensive stable isotopes available for purchase.

Of the remaining batches, an evaluation was made taking into account the cross-section of the isotopes and the gamma intensity and gamma energy of their corresponding activation products. The batches selected for initial evaluation for possible use in a taggant system are shown in Table 3.10. These batches were also selected because of their availability. Many of them were already purchased for other unrelated purposes.

Table 3.10: Initial selection of batches for the taggant system

Batch	Primary Isotope
155201	^{106}Cd
lx1440b	^{108}Cd
222101	^{114}Cd
201801	^{122}Te
209026	^{124}Te
ca297a	^{126}Te
166401	^{128}Te

3.4.4 Material Form

Batches can be purchased in a variety of chemical and physical forms. The price varies little between physical forms because the enrichment is the main cost in preparation of a batch. For cadmium and tellurium, the two common available

physical forms were metal and oxide powders. Tellurium metal (Te) powder was the least expensive tellurium form while cadmium oxide (CdO) powder was the least expensive cadmium form. Tellurium was purchased in pure metal powder form with a gravimetric factor of 1.0. Purchase of cadmium oxide powder necessitated the calculation of an individual gravimetric factor for every batch of cadmium because the atomic weight was different for each batch.

4. STARS AND BARS

Combining different nuclides together in differing amounts creates a vast number of possibilities that only grow with an increased selection of nuclides to choose from. To assess the utility of a taggant system, the maximum number of possible combinations is a critical metric. Combinatoric mathematics can be used to determine this metric if the total number of nuclides employed along with their associated enrichment increments are known.

4.1 Examples of Stars and Bars

4.1.1 *Purchasing Drinks*

One of the most difficult problems in purchasing drinks for a party is the sheer amount of combinations possible. Suppose there are five choices and a total of eight cases will be purchased to fulfill the needs of the guests. Multiple purchases of the same brand must be allowed as there are only five choices. In mathematical terms, each possible combination can be listed as a 5-tuple. The 5-tuple (1,1,2,3,1) would indicate one case of beverage A, one case of beverage B, two cases of beverage C, three cases of beverage D, and one case of beverage E. The problem could then be expressed as the possible number of different 5-tuples that can be formed to produce eight cases. This is the equivalent to determining how many non-negative integer solutions there are to

$$x_1 + x_2 + \cdots + x_r = n \tag{4.1}$$

or applied to the choice of particular beverage from the example above,

$$x_1 + x_2 + x_3 + x_4 + x_5 = 8. \quad (4.2)$$

The solution to this problem can be obtained through the use of a branch of combinatorics known as Stars and Bars [33]. The method of Stars and Bars is to visually divide the objects to be selected, stars, into different bins using bars. The 5-tuple (1,1,2,3,1) discussed earlier would be represented in stars and bars as $*|*|**|***|*$. There are eight stars which represent the eight cases that will be purchased and there are four bars that form the five bins which represent the five choices of beverages possible. The 5-tuple (4,0,4,0,0) can be represented in stars and bars as $****||****||$. In both 5-tuples, there are eight stars, 4 bars, and twelve characters total and this will be true of all 5-tuples that will determine the beverage selection for the gathering. There are 12 possible character positions that may be occupied by either a star or bar, but given the limitation that there must be 4 bars, the number of possible locations for the stars to occupy falls to only 8. Simple combinatorics can be used to determine the solution where n is the number of different beverage brands, or bins, $n - 1$ is the number of bars, and k is the number of cases that must be purchased, or stars:

$$\binom{n-1+k}{k} = \binom{bars+stars}{stars} = \binom{5-1+8}{8} = \binom{12}{8} = 495. \quad (4.3)$$

If the additional constraint is added that each bin must be populated with at least one star, or at least one of each type of beverage must be purchased, Eq. 4.3 is modified to

$$\binom{k-1}{n-1} = \binom{8-1}{5-1} = \binom{7}{4} = 35. \quad (4.4)$$

4.1.2 Distributing Spare Change

The same methods can be applied to just about any object. If there are seven coins that are to be divided amongst three people and each person gets at least one, Eq. 4.4 can be used to arrive at the solution of $\binom{7-1}{3-1} = 15$ possible ways to distribute coins. If each person does not get at least one coin, Eq. 4.3 is used to form $\binom{3-1+7}{7} = 36$ distributions.

4.2 Isotopic Stars and Bars

To apply the concept of Stars and Bars to an isotopic vector, the value of enrichment represented the stars and the number of isotopes in the isotopic vector represented the bars. The combinations of different isotopic vectors of an enriched element can be determined through the use of Stars and Bars.

4.2.1 Uranium Enrichment

Suppose there is a factory that produces enriched uranium, but each pass through their secret enrichment process increases the enrichment of ^{235}U in 5% increments. This factory begins the enrichment process with a pure feed material containing 5% enriched uranium ^{235}U and 95% ^{238}U . All other isotopes of uranium were removed prior to arrival at the factory. The special enrichment process produces completely depleted uranium as a waste, containing 0% ^{235}U and 100% ^{238}U and enriched uranium ^{235}U up to 100% ^{235}U . A list of enrichments capable of being produced by the factory is shown in Table 4.1.

The factory can produce 21 different enrichments for customers based on Table 4.1. If there is an additional requirement that both isotopes of uranium be present,

Table 4.1: Uranium enrichment increments

^{235}U Fraction (%)	^{238}U Fraction (%)
0	100
5	95
10	90
15	85
20	80
25	75
30	70
35	65
40	60
45	55
50	50
55	45
60	40
65	35
70	30
75	25
80	20
85	15
90	10
95	5
100	0

the number falls to 19, because pure ^{235}U and pure ^{238}U are not allowed. To apply stars and bars, the sample was divided into 20 equal parts of 5% enrichment each and each 5% increment of sample represented one star. The feed material containing 5% enriched ^{235}U would be represented as

*|*****.

Each star represents 5% enrichment and each bin represents either ^{235}U or ^{238}U . As the left bin only has one star, it signifies 5% ^{235}U . To represent 90% enriched ^{235}U , most of the stars are shifted to the ^{238}U bin,

*****|**.

With 18 stars at 5% each, the ^{235}U bin now represents 90% and with only 2 stars, the ^{238}U bin only represents 10%. In both cases, the total number of bins is two, the total number of bars is one, and the total number of stars is 20 since there is always 100% of sample present.

The possible number of combinations can be verified using Stars and Bars. Eq. 4.3 yields a value of $\binom{2-1+20}{20} = \binom{21}{20} = 21$ combinations. If both isotopes must be present, Eq. 4.4 applies to yield $\binom{20-1}{2-1} = \binom{19}{1} = 19$ combinations.

The idea of using Stars and Bars to determine the possible number of enrichment combinations can be applied to even finer enrichment schemes. If the sample was enriched in 1% increments, the number of stars would increase from 20 to 100, or $\frac{100}{\text{increment}}$ stars. If there were more than two isotopes present, the number of bins would increase to match the number of isotopes.

4.2.2 Multiple Elements

Combinations of isotopes from more than one element may be used. As long as the total number of stars sums to 100%, Stars and Bars will still be applicable. When an element with three isotopes is combined with an element with six isotopes, the elements can be treated as one element if the enrichment increments across all isotopes are the same. For 1% increments with 9 different isotopes, the number of combinations possible is $\binom{100-1}{9-1} = 171,200,862,756$.

4.2.3 Different Enrichment Increments

In practice, different isotopes will most likely be incremented at different intervals from each other. Very rare isotopes will most likely be incremented in minute quantities, possibly less than 1% of the overall sample, while other isotopes in the same sample will be incremented in large quantities such as 10% or 25%. For these different combinations of increments, a computer program was written to numerically

determine each possible isotopic vector given a list of increments. For the arbitrary isotopic vector and increments shown in Table 4.2, there were 531,388 possible combinations. Obviously, more isotopic vector combinations are possible with a larger number of isotopes and smaller increments for each isotope.

Table 4.2: Different enrichment increments with multiple isotopes

Isotope	Increment (%)
1	5
2	1
3	5
4	1
5	10
6	5
7	5

5. DETERMINATION OF PARTIAL BATCH CONTRIBUTIONS

5.1 Systems of Linear Equations

If NAA is performed on a single batch, the isotopic vector obtained through NAA should closely match that obtained from the initial MS assay of the batch. There will be errors associated with both the process of NAA and MS, but these errors should not significantly hinder this simple attribution as long as there is no other batch substantially similar in isotopic vector. If NAA is performed on a mixture of two or more batches, the fractional presence of individual batches, x_i , can be determined through the solution of a system of linear equations:

$$\begin{aligned} a_{11}x_1 + a_{12}x_2 + \cdots + a_{1j}x_i &= b_1 \\ a_{21}x_1 + a_{22}x_2 + \cdots + a_{2j}x_i &= b_2 \\ &\vdots \\ a_{i1}x_1 + a_{i2}x_2 + \cdots + a_{ij}x_i &= b_j \end{aligned} \tag{5.1}$$

where,

a_{ij} is the atomic fraction of the j^{th} isotope present in the i^{th} batch;

x_i represents the fractional presence of batch i in the whole sample; and

b_j is the measured atomic fraction of isotope j present in the whole sample.

5.2 Matrix Form

Eq. 5.1 can be formalized into standard matrix form, $Ax = b$, as

$$\begin{bmatrix} a_{11} & a_{12} & \dots & a_{1j} \\ a_{21} & & \ddots & \\ \vdots & & & \\ a_{i1} & & & a_{ij} \end{bmatrix} \begin{bmatrix} x_1 \\ \vdots \\ x_i \end{bmatrix} = \begin{bmatrix} b_1 \\ \vdots \\ b_j \end{bmatrix} \quad (5.2)$$

where the objective is to solve for the x vector. The b vector is formed by the isotopic vector of the entire sample as a whole, as measured through NAA, or other isotopic method such as MS. The A matrix is simply formed from known assay information of the stable isotope batches that are known to be present in the measured sample. This information is usually gained through MS upon purchase of the stable isotope batches. The assay information of the batches is required to translate the isotopic vector and is the hidden key to obtaining the taggant vector used for attribution. Table 3.4 or Table 3.6 could form the A matrix if the batches in those tables were used.

5.2.1 Necessary Properties of the A Matrix

To form a well-posed problem with a unique solution, A must be a square matrix, ensuring that the number of isotopes is equivalent to the number of batches present to allow accurate attribution to a particular batch. This does not mean that each isotope must be present in every batch, as null values are valid in the A matrix. This requirement only specifies that the isotopes measured through NAA must be present in at least one batch and that there is at least one batch for every isotope that is measured. Extraneous isotopes of many different elements will be measured because of contamination but these isotopes will not be used for the fractional determination of batches.

5.2.1.1 *Diagonal Dominance*

While it is clear that the use of only 100% pure batches will allow almost trivial fractional batch attribution, the process becomes more difficult as the samples move away from purity. Through inspection of A in the context of solving a system of linear equations to determine the fractional presence of different batches, it can be shown that A must be diagonally dominant [34]. The Levy-Desplanques theorem extends this to ensure that diagonal dominance will also ensure that A is invertible [35] and non-singular. This implies the previously mentioned constraint that A is a square matrix, allowing for a well-posed problem, and being invertible, leading to a unique solution. Diagonal dominance implies that the isotopic vector of each batch will be significantly different from each other. This criterion requires that the primary isotope of each batch retains a majority share when combined with other batches. A batch containing greater than 50% enrichment of the primary isotope will fulfill this requirement. The introduction of an additional constraint that all values of A are non-negative is also necessary to form a lower boundary of the isotopic vector because enrichment values must fall between 0% and 100%.

5.2.2 *Solution Methods*

The solution to Eq. 5.2 can be obtained through a variety of solution methods [36], usually involving a direct method, as opposed to an iterative method, that will yield the x vector in a straightforward manner. For the determination of isotopic vectors, direct methods are generally sufficient as iterative methods are reserved for very large or sparse systems. The solution of the x vector represents the fractional presence of a batch in an entire sample analyzed through NAA or other similar means.

5.3 Matrix Error

There will be uncertainties in NAA measurement of the sample along with errors associated with the initial assay of the batch obtained through MS. Both of these errors should be small enough not to preclude the positive identification of a small number of batches and isotopes. As the number of batches and isotopes increases, this error will also increase. Terms for error are added to Eq. 5.2 to form Eq. 5.3 [36, 37, 34, 38, 39] for a system of dependent equations:

$$\frac{\|\delta x\|}{\|x\|} \leq K(A) \left(\frac{\|\delta b\|}{\|b\|} + \frac{\|\delta A\|}{\|A\|} \right) \quad (5.3)$$

where,

δb is vector containing the error values of b ;

δA is the matrix containing the error values of A ; and

$K(A)$ is the condition number of A .

The δb vector is formed by the experimental error associated with NAA measurement and the δA matrix is formed from the error of the initial assay of the batches, such as examples given in Table 3.5 or Table 3.7.

5.3.1 Condition Number

The condition number, K , of a matrix is a measure of how invertible the matrix is and will also determine how sensitive x is to changes in A and b . A matrix that is not sensitive to changes in A and b would have a condition number of 1, such as the identity matrix. A singular matrix that is not invertible will have a determinant of 0 and a condition number of ∞ , such as a matrix where all elements are 1. A matrix with a large condition number is sensitive to changes in A and b such that

small changes, or error, in either the NAA measurement or MS assay will cause great changes in x . A condition number of 1 will indicate proportional change in x to a change in either A or b .

5.3.1.1 Calculation of Condition Number

The condition number is determined by

$$K(A) = \|A\| \cdot \|A^{-1}\| \quad (5.4)$$

where,

A^{-1} is the inverse of A .

The condition number is dependent upon choice of norm used. The Frobenius norm [39], shown in Eq. 5.5, and the Euclidean norm, shown in Eq. 5.6 are adequate choices:

$$\|A\|_F = \sqrt{\sum_{i=1}^m \sum_{j=1}^n |a_{ij}|^2} = \sqrt{\text{tr}(A^*A)} = \sqrt{\sum_{i=1}^{\min\{m,n\}} \sigma_i^2} \quad (5.5)$$

where,

A^* is the conjugate transpose of A ; and

$\text{tr}(A) = \sum_{i=1}^n a_{ii}$, the sum of elements along the main diagonal of A .

For isotopic vectors containing only real numbers and no complex values, the conjugate transpose is equivalent to the transpose such that, $A^* = A^T$:

$$\|A\|_2 = \sqrt{\lambda_{\max}(A^*A)} \quad (5.6)$$

where,

λ_{\max} is the maximum singular value, or largest eigenvalue, of A^*A . As with the Frobenius norm, the simple transpose may be substituted for the conjugate transpose for all isotopes that are not imaginary.

To find the maximum singular value, a power iteration can be used to solve

$$Ax = \lambda A. \tag{5.7}$$

Iterative refinement of Eq. 5.8 is performed with the replacement of x by λ during each iteration until convergence is achieved:

$$\lambda = xAA^{-1}. \tag{5.8}$$

The Frobenius norm is always as large as, and usually larger than, the spectral radius obtained through use of the Euclidean norm. The Frobenius norm would be similar to taking the variance of two additive numbers as the sum of the absolute values of the variance instead of the square root of the sum of the squares of the variances, which would be akin to the Euclidean norm. In this way, the Frobenius norm will tend to overestimate the variance while the Euclidean norm will tend to serve as a more accurate representation of the variance.

6. TAGGANT SYSTEM

6.1 Taggant Vector Identification

6.1.1 *Isotopic Vector to Taggant Vector*

The resulting taggant vector obtained in Eq. 5.2 is used as the basis of a taggant system. A taggant system is one in which items or classes of objects are marked or tagged with a combination of batch material to form a unique and particular isotopic vector. This material may be recovered and analyzed using NAA to form the isotopic vector and then translated to the taggant vector using Eq. 5.2. This taggant vector is then correlated with a unique taggant identifier that is stored in a database during taggant manufacture. Upon recovery of the item, the isotopic vector is determined through NAA and then translated to the taggant vector. Correlation with a taggant vector in the stored database will yield the identity of the item.

6.1.2 *Taggant Vector Storage in a Database*

The translation of taggant vector to database identifier is flexible and may be changed depending upon database storage requirements and other factors which depend upon the user and particular application requirements. In general, the unique taggant identifier will need to have adequate length to reflect the uniqueness of each taggant. For taggant systems with a small number of possible combinations, the taggant identifier will have a short length. As the number of possible combinations grows, the length of the identifier will also need to increase to uniquely identify all possible taggant combinations.

A suggested method to store the taggant vector in a database was arbitrarily chosen as a conversion of the taggant vector to basis points. This storage method

assumes that the error associated with NAA, manufacture, and measurement of the taggant will always exceed a resolution of 0.01% and there is no need to create a database with more precision than 1‰. This level of error was deemed to encompass all possible error and that all taggants would have measured error of at least 1‰. The number of digits is arbitrary and may be added or reduced. A shorthand system may use per mil instead of basis points if it is known that the error will always exceed 1‰.

With the conversion to basis points, each nuclide present in the taggant will represent 5 digits and will be concatenated to form a unique number with the length corresponding to the number of nuclides. A taggant vector with 4 nuclides each representing 25% of the whole will form a 20 digit ($5 \times 4 = 20$) identifier which would be "02500025000250002500" where each segment "02500" would represent the fraction of each isotope expressed in basis points. The order of each segment would correspond to a particular nuclide. This order would be determined at manufacture and would be used across the whole taggant system for uniformity. If a particular nuclide is not present in a taggant at manufacture, the corresponding segment would continue to be present but would represent the absence of the nuclide as "00000". As there will always be a combined sum of 100%, the sum of all segments in the identifier must always sum to 10000‰.

6.1.3 Comparison Against the Original

It is envisioned that a complete tagging system will record the unique taggant vector and also retain a small sample of manufactured taggant to be used later for comparison. After receipt of an unknown material, the taggant vector will be determined. An additional comparison to a stored reference material will lend credence to the authenticity of such an attribution, especially if comparative NAA is used.

6.2 Vector Phase Space

6.2.1 Summation to 100%

The taggant vector will undoubtedly occupy n-dimensional phase space corresponding to the number of nuclides present in a taggant. For taggants containing only one nuclide, the phase space will be 1-dimensional and the vector will have a magnitude between 0% and 100%, with the length corresponding to the fractional presence of the particular nuclide. A taggant system utilizing 1-dimensional phase space is physically impossible though as a balance nuclide must be present to make up the fraction of sample that is not occupied by another nuclide, such as the 2 isotopes of uranium shown in Table 4.1. A large number of different nuclides will yield a larger number of possible combinations and each additional nuclide will add one more dimension to the taggant vector. Each unique vector will occupy a particular voxel in n-dimensional phase space. The maximum magnitude of a particular vector in each dimension is determined by Eq. 6.1 and illustrated in Table 6.1 assuming that vectors along each dimension have a length between 0% and 100% to represent the real fraction that each nuclide may constitute of the whole taggant. Maximum vector lengths in Table 6.1 were calculated by measuring the maximum Euclidean distance between any two vertices:

$$D = \sqrt{N \times 100\%^2} \quad (6.1)$$

where,

D is the maximum Euclidean distance possible in N dimensional space.

While mathematically, the summation of elements of a vector in n-dimensional phase space may exceed a total of 100%, this is not physically possible as the sum-

Table 6.1: Maximum length of vector in n-dimensions

N	D (%)
1	100.0
2	141.4
3	173.2
4	200.0
5	223.6
6	244.9
7	264.6
8	282.8
9	300.0
10	316.2

mation of the isotopic vector and corresponding taggant vector must always be equal to 100% to represent the physical idea that all nuclide fractions in all physical matter must always sum to 100%. The physical solution space for a taggant containing 3 nuclides would fall along the surface of a plane with vertices at 100% in the first octant. All possible physical combinations of the 3 nuclides must fall on the surface of the plane, as there is no physical way to combine 90% of nuclide 1, 50% of nuclide 2, and 40% of nuclide 3. The sum of 180% is nonphysical and would not fall on the surface. The trivial case of 0% for all nuclides is acceptable physically, meaning there is no material present. It would create a vector of 0 length at the origin, but is not acceptable mathematically because it does not fall along the solution plane and would represent no taggant present.

6.2.2 Vector Normalization

The error associated with manufacturing and measurement of each taggant will cause the solution vector to deviate from the solution plane as the measured vector will not sum to exactly 100%. This means that the solutions will tend to fall along

the surface, but not exactly on it. To measure the perceived difference between two taggant vectors, the Euclidean distance was calculated with a Euclidean norm and normalized to 1-dimensional phase space by dividing the vector magnitude by the corresponding maximum phase space of the vector dimension as shown in Table 6.1. This allowed a comparison of vectors across many different dimensions and normalized the resulting vector lengths, and associated errors, to a magnitude between 0 and 100 for all dimensions. This value represents the fraction of the maximum possible vector magnitude achievable in any particular dimension, including unrealistic vectors. While the true Euclidean distance is preferable to measure differences in similar isotopic vectors of similar dimension, this normalized value serves as an approximate comparison across different taggant systems with varied number of nuclides. The greatest error in all dimensions is then normalized to 100% allowing easy comparisons of relative error across different dimensions.

6.3 Gharv Computer Program

The Gharv program was written and used to perform the calculations and translations necessary to implement a taggant system. The complex and varied calculations were carried out using this program, providing an efficient and timely means of assessing and identifying taggants. Gharv was originally written as a general purpose program to aid in common calculations pertaining to NAA. The engine on which the program is built is the inverse calculation of originating stable nuclide mass gleaned from information about activation product activity, which is useful in the NAA community to quickly determine concentrations and masses of unknown materials after irradiation. Selections of the program are located in Appendix A.

6.3.1 *Inverse Calculation of the Stable Nuclide Atomic Fraction*

Using a library of applicable nuclear data, such as cross section, half-life, and activation pathway, the activation product radioactivity information obtained from gamma-ray spectroscopy is entered into the program along with the radionuclide identified by characteristic gamma energies. Assuming no branching activation and that the activation product was created by the irradiation of one isotope, the program can be used to calculate the initial amount of target nuclei using Eq. 2.4 with the error expressed in Eq. 2.6. If a reference sample is available and NAA is performed on this reference to obtain radioactivity information, then Eqs. 2.7 and 2.8 are used instead. At the completion of this step, the atomic fraction of each initial stable nuclide is known, forming the basis of the isotopic vector.

6.3.2 *Translation to Taggant Vector*

The isotopic vector is then used to form the b vector in Eq. 5.2. The A matrix is populated by the isotopic vectors of each stable isotope batch. These vectors are obtained from MS information that is included with the purchase of each stable isotope batch. Solution of Eq. 5.2 yields the x vector, which is the taggant vector. The taggant vector is the fraction of each whole taggant that each stable isotope batch constitutes. Mixing 2 batches together equally would yield a taggant vector of 50% and 50%. The corresponding isotopic vector would not necessarily be 50% though but would most likely be near 50%. For all batches that are enriched to near unity in the primary isotope, the isotopic vector and taggant vector are not expected to significantly differ, but will differ.

6.3.3 *Euclidean Distance of Taggant Vectors*

The initial mass of each batch manufactured into a taggant is entered into the program and is then translated to an isotopic vector. This vector is then translated to a manufactured taggant vector, which serves as the unique taggant identifier. After formation of the measured taggant vectors through NAA, the Euclidean distance is calculated between the manufactured and measured taggant vectors and normalized to 100. This value gives a reasonable error estimate of the taggant system. A value of 0 indicates that there is no error and the measured unknown taggant is exactly equivalent to the known manufactured taggant. A value of 100 indicates that the error is so great that the unknown taggant is as dissimilar to the known taggant as possible. A high error may also indicate that the solution vector has exceeded physical boundaries, resulting in a taggant vector with a drastic departure of total summation away from 100%. For purposes of calculation of the Euclidean distance to determine the error, the obviously incorrect measured taggant vectors were used. Once the Euclidean distance had been ascertained, the taggant vectors were then normalized to 100% by assuming that the error was evenly distributed proportionally relative across all nuclides so that each nuclide bore a similar share of the combined error across the isotopic vector. This was done to reflect the physical requirement that all isotopic vectors must sum to 100% and as an extension, all taggant vectors must also sum to 100%.

6.3.4 *Nearest Neighbor Search*

The Gharv program uses a generalization of the nearest neighbor search algorithm [40, 41] to classify unknown taggants. After the unknown material is processed and measured, the resulting taggant vector is compared against the database of manufactured taggants. Taggants are selected from the database that have similar

isotopic vectors to the measured taggant. After the selection of candidate taggants are chosen from the database, the Gharv program can be used to calculate the Euclidean distance between the measured taggant and all candidate taggants. The possible taggants are listed in order of increasing Euclidean distance with the most likely taggant candidates at the top of the list.

7. REACTORS AND FACILITIES

7.1 Facilities Located at Oak Ridge National Laboratory

7.1.1 *High Flux Isotope Reactor*

While also home to the second oldest reactor in the world, the X-10 Graphite Reactor, Oak Ridge National Laboratory's flagship reactor is the High Flux Isotope Reactor (HFIR). Designed by graduate students [42] prior to initial criticality in 1965, the reactor is a vital component in the production of transuranic elements for medical, commercial, and research purposes. The most notable of these elements, ^{252}Cf , is supplied by HFIR to the western world for cancer therapy, materials testing, and the detection of explosives in luggage.

Operating at 85 MW_{th} , HFIR is a light water reactor employing a flux trap design in which the annular core surrounds a central region, as shown in Fig. 7.1. Thermal neutrons are used for irradiation of samples in the central region for long periods of time. A beryllium reflector surrounds the core to enhance neutron economy and provide valuable experimental facilities.

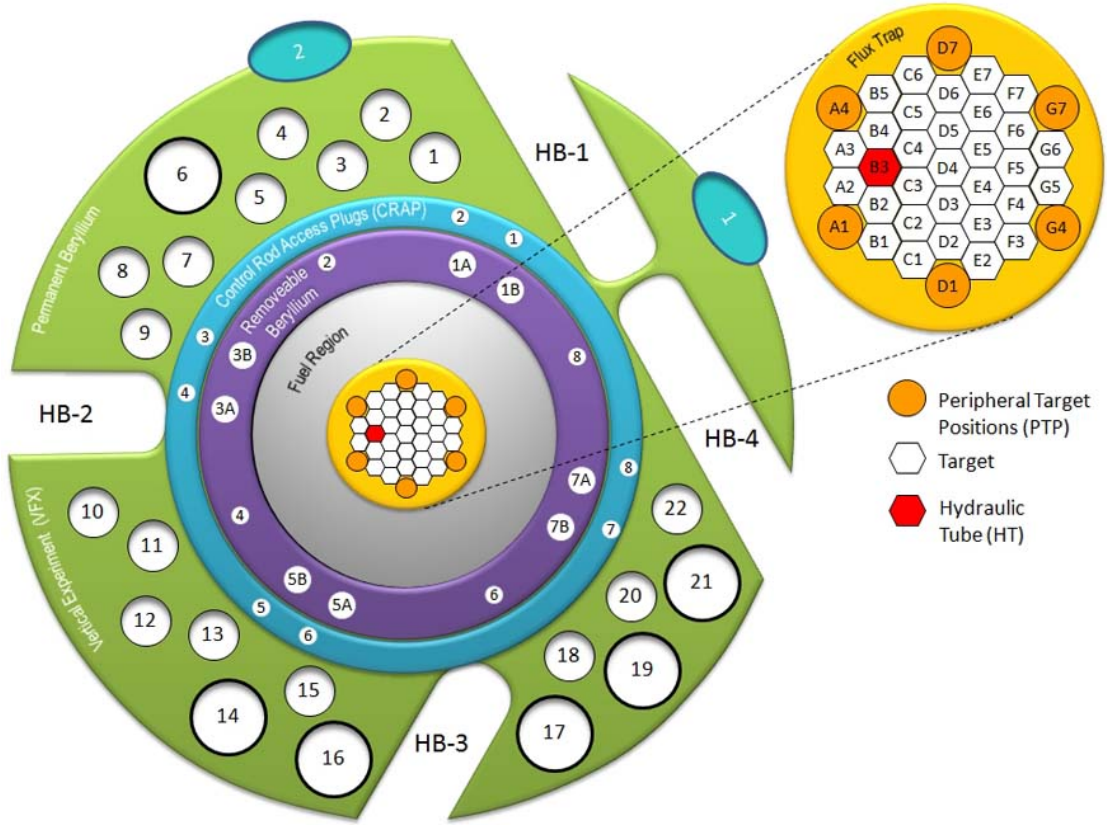


Figure 7.1: Layout of the HFIR, courtesy of Oak Ridge National Laboratory, U.S. Dept. of Energy.

7.1.2 Neutron Activation Analysis Lab

7.1.2.1 Overview of Irradiation Process

For short irradiations, a pneumatic facility using pressurized gas quickly transports small samples between the Neutron Activation Analysis Laboratory (NAAL) and position Vertical Experiment-7 (VXF-7), shown in the permanent beryllium region in Fig. 7.1. This position is located in the beryllium reflector region of the reactor and has a thermal neutron fluence rate [5] of $4.0 \pm 0.2 \times 10^{14}$ neutrons $\text{cm}^{-2} \text{s}^{-1}$ and a resonance region neutron fluence rate of $1.0 \pm 0.2 \times 10^{13}$ neutrons $\text{cm}^{-2} \text{s}^{-1}$.

s^{-1} . A pressure transducer provides a signal to start the irradiation timer when the sample arrives in the irradiation position and begins to block the continuous flow of gas. For samples that are anticipated to be very radioactive, a hold station is available to delay the return of the sample. Instead of returning to the lab, the sample pauses at a point outside of the reactor, yet still in the reactor pool to allow safe radioactive decay. The pneumatic system in the NAAL discharges into a hot cell equipped with remote handling arms and shielded glass. When the sample is radiologically safe for removal from the hot cell, it is passed through an airlock into the lab. A radiological control technician (RCT) surveys the sample to check for unsafe levels of radiation. Samples may then be returned to the hot cell for further decay if needed. If the sample is deemed acceptable by the RCT, it is taken to a fume hood. The sample material is removed from the graphite rabbit, repacked to prevent contamination, and then counted on one of the many high purity germanium (HPGe) detectors available.

7.1.2.2 Packaging and Repacking of Samples

Samples to be irradiated for long periods of time in the reactor are packaged in a graphite container, or rabbit, that can withstand the harsh neutron environment of the reactor without deformation or melting that could lead to possible loss of radionuclides and contamination of the pneumatic system. The graphite rabbit has an internal diameter of 10 mm and an internal height of 18 mm and is shown in Fig. 7.2. The lid is attached and removed using a screwdriver to allow safe handling and repacking after irradiation and to reduce the radiation dose. The time needed to repack a sample after removal from the hot cell is generally 10 minutes.

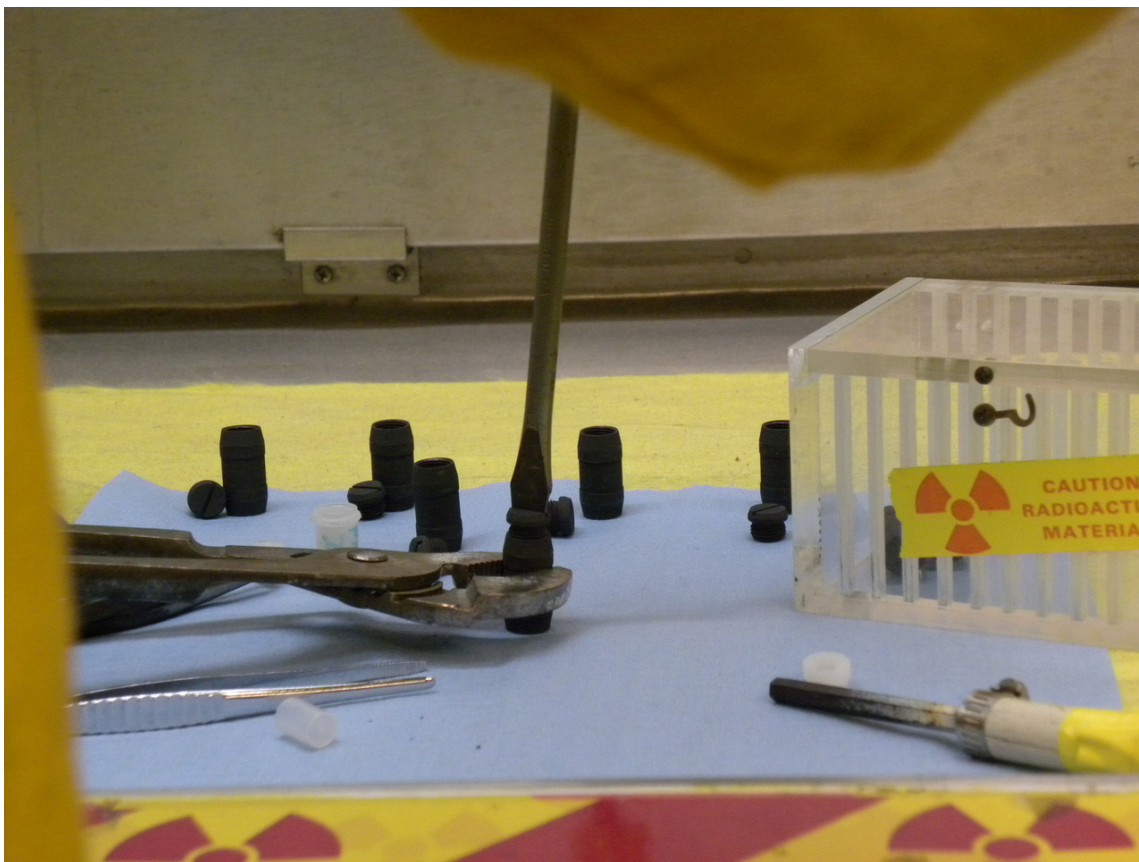


Figure 7.2: Repacking a graphite rabbit after irradiation.

7.1.2.3 Radioactivity Assay

After samples are removed from the graphite rabbit, they are placed in a clean polyethylene vial to prevent possible contamination of the counting detector. Samples irradiated in the HFIR reactor are assayed for radioactivity using a high purity germanium detector, model GC4520, manufactured by Canberra Industries with a relative efficiency of 45% and subsequent gamma-ray spectra are analyzed with Genie-2000 software, version 3.1 [43]. Lead shielding placed around the detector is used to inhibit cosmic and background radiation. The detector is also enveloped by a continuous stream of nitrogen gas to create a positive pressure environment. This

purges radioactive gases that originate from the the nearby reactor facility from the detector area. Detectors are frequently calibrated for both energy and efficiency. Background counts are used to compensate for nuclides that are persistently present during the counting process, specifically ^{41}Ar that is continuously produced in the pneumatic system piping.

7.2 Facilities Located at Texas A&M University

7.2.1 *TRIGA Mark I Reactor*

While also home to the second oldest reactor still operating at a university, the AGN-201M reactor, Texas A&M University's flagship reactor is the TRIGA Mark I, located at the Nuclear Science Center. Reaching initial criticality in December of 1961, the reactor has a maximum steady state power of 1 MW_{th} along with pulsing capability. With multiple irradiation facilities, the reactor is useful for commercial and experimental irradiations. For short irradiations, a pneumatic system is employed to access location D2 in the core with a thermal neutron fluence rate of 1.04×10^{13} neutrons $\text{cm}^{-2} \text{ s}^{-1}$ and a resonance region neutron fluence rate of 2.4×10^{11} neutrons $\text{cm}^{-2} \text{ s}^{-1}$. Samples are pushed by CO_2 through tubing to reach the core and are quickly ejected from the core at the end of irradiation and returned promptly to the NAA laboratory.

7.2.2 *NAA Laboratory*

7.2.2.1 *Packaging of Samples*

Samples are packaged in polyethylene vials and sealed using a soldering iron by melting the plastic under slight negative internal pressure, shown in Fig. 7.3. This allows gaseous expansion in the vial during irradiation. With an internal diameter of 14 mm and an internal height of 53 mm, the rabbit used at Texas A&M University

is significantly larger than the rabbit used at Oak Ridge National Laboratory.



Figure 7.3: Polyethylene vial used for irradiation at Texas A&M University.

Smaller polyethylene vials may be placed inside the rabbit if desired. The internal diameter is slightly smaller at 10 mm while the internal height is less than half that of the rabbit at 22 mm. This allows a snug fit of 2 small vials into each rabbit.

7.2.2.2 Operation of Pneumatic System

The pneumatic system is operated from a small hood, which is shown in Fig. 7.4. Samples are loaded into the pneumatic system and inserted into the reactor through the use of a control panel just outside the hood. Samples are returned automatically

when the timer on the control panel has expired, which can be set to a maximum of 999 seconds or slightly longer than 16 minutes.



Figure 7.4: Pneumatic system hood where samples are loaded into the reactor.

Upon return of the sample, a brief gross count is used to check for unsafe levels of radiation. Sample rabbits are then wiped clean to remove possible contamination on the outside of the rabbit and then placed in a plastic bag. Opening of the rabbits is not necessary. The sample can then be counted on a variety of HPGe detectors. The time between sample removal from the core and the beginning of counting is usually less than 5 minutes because there is no need to repack the irradiated sample

in a different container for counting.

7.2.2.3 Counting Laboratory

Samples irradiated in the TRIGA reactor are counted using a high purity germanium detector, model GC3518, manufactured by Canberra Industries with a relative efficiency of 35% and subsequent gamma-ray spectra are analyzed using Genie-2000 software, version 3.1a [44].

8. SOLID PHASE

8.1 A Select Class of Reactor

The HFIR was chosen to test the proposed taggant system. The choice of HFIR was based upon the unusually high neutron fluence rate of the reactor that is only reached among a select group of research reactors in the world. Designing a system with the requirement of a high neutron fluence rate limits the ability to detect the taggant and would hinder discovery of the taggant and unauthorized use. The high fluence rate also increases the chances of detecting nuclides of small abundance, allowing a greater variety of taggant vectors.

8.2 Solid Standards

8.2.1 Preparation

Standards of natural elements were employed to allow the added advantages of comparative NAA in addition to parametric NAA. The masses of the standards are shown in Table 8.1. Each standard was composed of two foils containing natural cadmium and tellurium metal (abundances are shown in Table 3.1).

Table 8.1: Masses of solid cadmium and tellurium standards

Element	Mass (mg)
Solid Standard 1	
Cadmium	1.74
Tellurium	0.46
Solid Standard 2	
Cadmium	1.30
Tellurium	0.36

8.2.2 Comparative Results

Irradiation of the natural standards proved inadequate for use as a comparative standard for the taggants. The low natural abundance of ^{106}Cd , ^{108}Cd , and ^{124}Te prevented use of the natural foils as a standard because no activity was reported for these radionuclides. The solid standards were instead compared to each other using the comparator method with the common isotopes that were reported for both standards. Using Eq. 2.7, the mass of the solid standard 2 foils were calculated using the measured activities of both standards along with the known mass of solid standard 1. This value was then compared to the known mass of solid standard 2. The results are shown in Table 8.2 according to the isotope activity used for calculation. If there were no errors, the values in Table 8.2 would match those of Table 8.1.

Table 8.2: Solid Standard 2 mass calculated with comparator method

Isotope	Mass (mg)
Cadmium	
^{115}Cd	0.951
Tellurium	
$^{123\text{m}}\text{Te}$	0.369
^{127}Te	0.392
$^{131\text{m}}\text{Te}$	0.336

The cadmium and average tellurium masses of Table 8.2 are 0.951 mg and 0.364 mg, respectively. The measured solid standard 2 cadmium mass of 1.30 mg is significantly higher than the calculated mass of 0.951 mg, but such error is expected when only one isotope is used for calculation. The average NAA measured solid standard 2

tellurium mass of 0.364 mg, differing only by 4 μg , shows the advantage of the comparator method when multiple isotopes are used for comparison.

8.3 Preparation of the Solid Phase

Stable isotopes are packaged and shipped in solid powder form, which is shown in Fig. 8.1. Batches were mixed together directly in powder form for irradiation. The staff at the HFIR, like many research reactors, has a policy to greatly restrict the irradiation of liquid or gaseous forms because they are more likely to expand under intense gamma irradiation and high temperature. This expansion may cause failure of the rabbits and contamination of the entire pneumatic system.

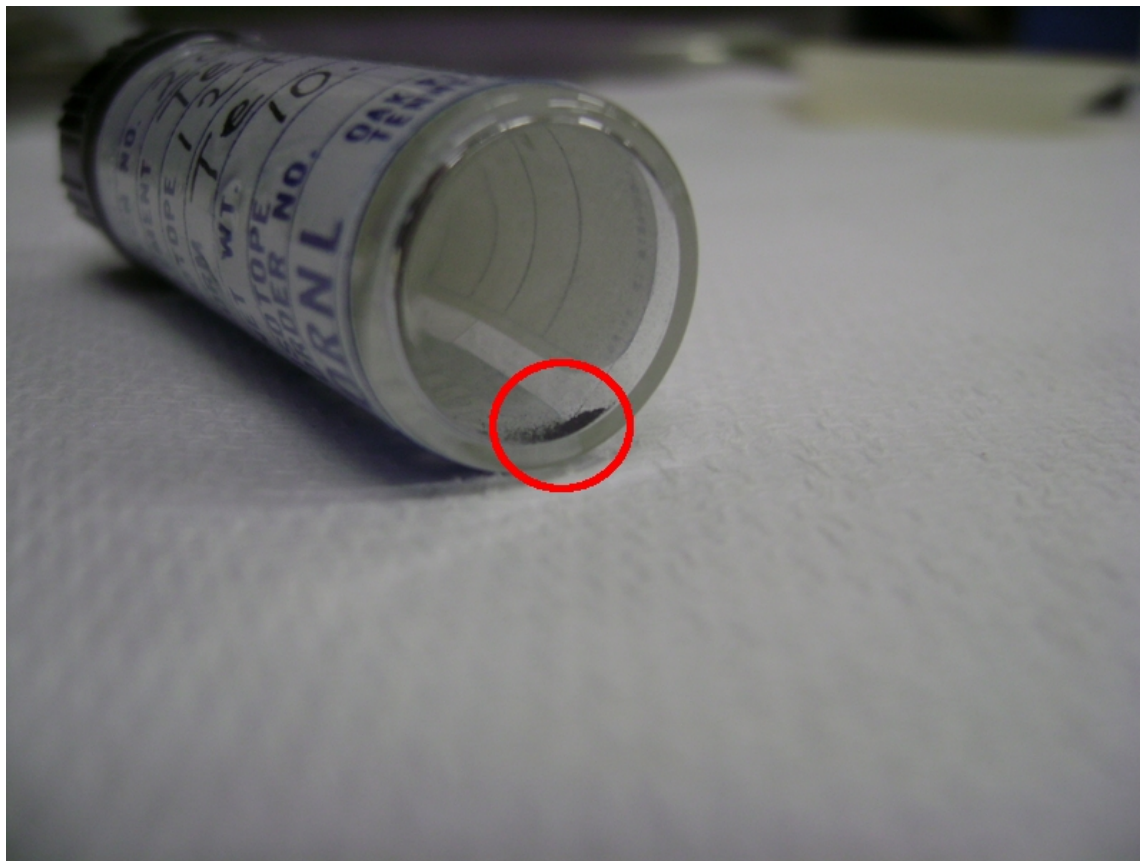


Figure 8.1: Shipped stable isotope in original packaging.

8.3.1 Irradiation Parameters

8.3.1.1 Isochain

The computer code Isochain [45] was employed to calculate the buildup and decay of different nuclides during irradiation as a guide to the selection of mass values for each batch. The batches selected for irradiation are shown in Table 3.10. The optimal mass for irradiation of each batch was deemed that at which the resulting activation product radioactivity stemming from a batch was adequate for NAA, which is approximately 370 kBq. These masses are shown in Table 8.3.

Table 8.3: Optimal masses of batches obtained using Isochain

Batch	Primary Isotope	Optimal Mass (mg)
155201	^{106}Cd	0.25
lx1440b	^{108}Cd	1.0
222101	^{114}Cd	0.05
201801	^{122}Te	0.10
209026	^{124}Te	1.50
ca297a	^{126}Te	0.1
166401	^{128}Te	0.75

8.3.1.2 Irradiation Time

Irradiation time was chosen to be 1 hour. A longer irradiation time allowed more efficient use of limited material resources but was still short enough to avoid experimental failure from embrittlement. The choice to irradiate for 1 hour was also guided by previous irradiations of enriched CdTe in the NAAL.

8.3.1.3 Decay Time

To allow most short-lived unimportant trace elements time to decay, a rabbit is usually held in the pool decay station for at least a few minutes before it is manually returned to the hot cell, where it may then be moved to the hood for repackaging, and then to the detector. As all sample handling is completed in accordance with federal regulations [46, 47], each sample must be surveyed upon removal from the hot cell as shown in Fig. 8.2.



Figure 8.2: A radiological control technician performs a survey upon removal from the hot cell.

RCTs are not assigned to a particular location or laboratory and are subject to the business needs of HFIR, which change daily while irradiations must be scheduled far in advance. To ensure that a RCT was available to extract the sample from the hot cell before the decay of key isotopes took place, a decay time of 24 hours was chosen. This required a larger amount of material to be irradiated per sample but almost guaranteed that a RCT would be available at some point for extraction.

8.3.1.4 Counting Time

A counting time of 1 hour was arbitrarily chosen because of scheduling constraints of the detectors of the NAAL. The detectors in use at the facility are in constant demand and 1 hour was deemed adequate without disrupting NAAL operations. A longer counting time was not necessarily warranted as a taggant system would need relatively rapid analysis.

8.3.2 Weighing

The balance used for all mass measurements pertaining to samples irradiated in HFIR was a Mettler Toledo model AB135-S/FACT Classic Plus balance. It is a dual range scale with readability ranges of 100 μg and 10 μg and is located on a marble table, seen in Fig. 8.3. A plastic boat for each batch was placed on the scale and then tared. Batches were poured into the boat to achieve a mass as close as possible to the calculated optimal mass for each batch. After weighing, all batches were combined in a high purity quartz ampoule. The masses of batches indicated by their primary isotopes are shown in Table 8.4.

Table 8.4: Masses of batches for solid taggants

Batch	Mass of Taggant 1 (mg)	Mass of Taggant 2 (mg)
^{106}Cd	0.69	1.03
^{108}Cd	0.75	1.05
^{114}Cd	0.64	0.05
^{122}Te	0.20	0.11
^{124}Te	0.62	0.68
^{126}Te	0.37	2.61
^{128}Te	0.32	1.37
Total	3.59	6.90



Figure 8.3: Marble table and balance.

The mass values of each batch, along with the isotopic vector of each batch were

processed using the Gharv program to produce a set of taggants that were unique to each sample based on the isotopic and taggant vectors. The taggant vectors are shown in Table 8.5. These taggants would form the database of such a system and be used for comparison should an illicit sample be recovered at a later time. The taggant vectors for the solid phase taggants, expressed in basis points are shown in Table 8.6.

Table 8.5: Solid phase taggant vectors

Isotope	Taggant 1 (%)	Taggant 2 (%)
^{106}Cd	19.65	15.27
^{108}Cd	20.95	15.27
^{114}Cd	17.21	0.7
^{122}Te	5.72	1.64
^{124}Te	17.46	9.97
^{126}Te	10.26	37.68
^{128}Te	8.74	19.48

Table 8.6: Solid phase taggant vectors expressed in basis point (‰)

Taggant	^{106}Cd	^{108}Cd	^{114}Cd	^{122}Te	^{124}Te	^{126}Te	^{128}Te
1	01965	02095	01721	00572	01746	01026	00874
2	01527	01527	00070	00164	00997	03768	01948

8.3.3 Quartz Ampoules

Synthetic high purity quartz ampoules were used to reduce production of impurities and to contain the irradiated material as required by safety guidelines. The

ampoules formed an extra barrier in case of experimental failure or melting. Using these ampoules also made removal of the potential contaminants from the graphite rabbit after irradiation easier. A tube of high purity quartz was cut and shaped by the Oak Ridge National Lab glass shop to form ampoules designed to be used in graphite rabbits that are used for irradiation. With a 6 mm outer diameter, the quartz ampoules were just large enough to prevent the loading of two ampoules into a single rabbit. After formation by the glass shop, the ampoules were all washed with concentrated nitric acid and rinsed with deionized water having a resistivity of 18 M Ω cm. Loose material powder was poured into the quartz ampoules from the plastic boats. The ampoules were returned to the glass shop for sealing, as seen in Fig. 8.4. The samples were then taken to the NAAL for placement into the graphite rabbits, leak testing, and placement on the docket for irradiation.

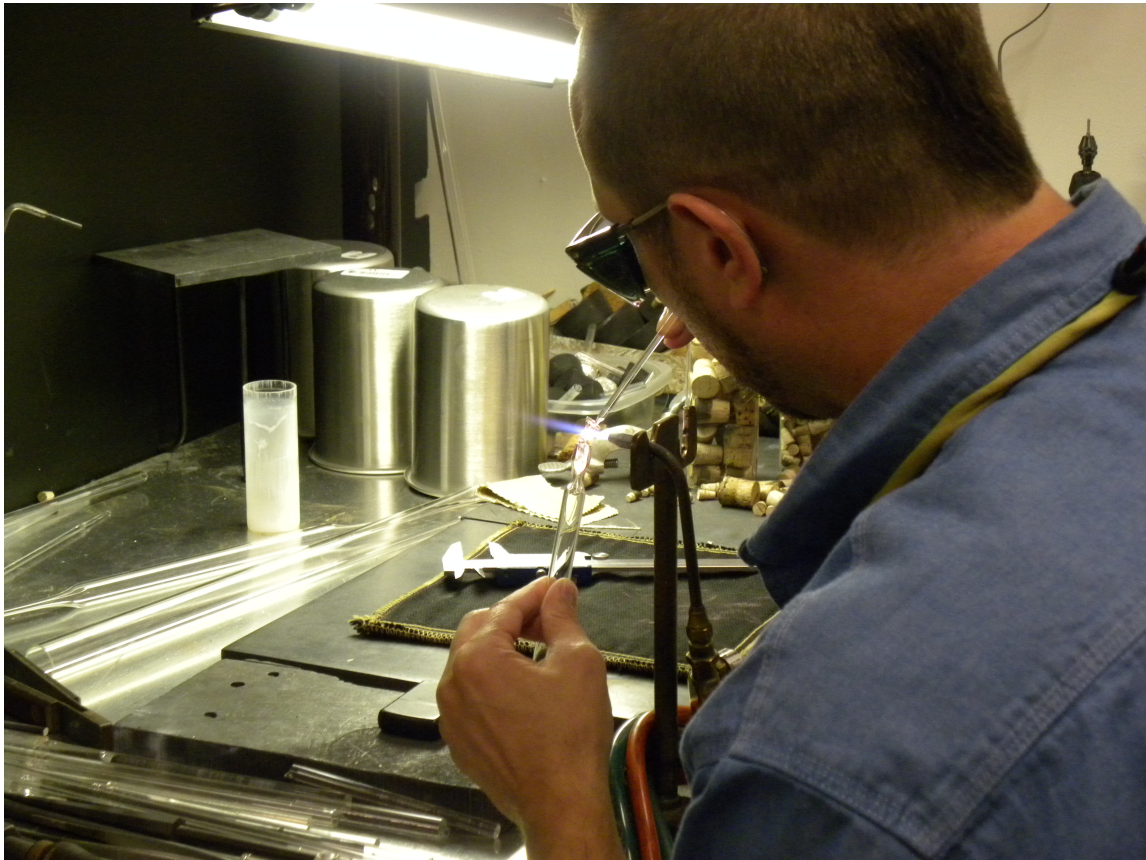


Figure 8.4: Sealing of a quartz ampoule at the Oak Ridge National Lab glass shop.

8.4 Counting of Solid Phase

Irradiation of the samples was completed in 1 day to minimize changing reactor core conditions arising from fuel depletion. The quartz ampoules were removed from the graphite rabbits and placed in clean plastic vials for counting, shown in Fig. 8.5.



Figure 8.5: A quartz ampoule is removed from a graphite rabbit and placed in a clean vial for counting.

The counting vials were labeled and counted on a HPGe detector at a vertical distance of 48 cm from the detector surface using a stand. The detector is shown in Fig. 8.6. Using Genie 2000 software [43] and a modified nuclear library updated with accurate values from Section 3, the gamma-ray spectra were processed to obtain the activities of the radionuclides at the end of irradiation (EOI). These EOI activity values were processed using the Gharv program to produce the measured taggant vectors. These vectors were compared to the prepared vector values to assess the Euclidean distance of the prepared and measured taggant vectors of each sample.

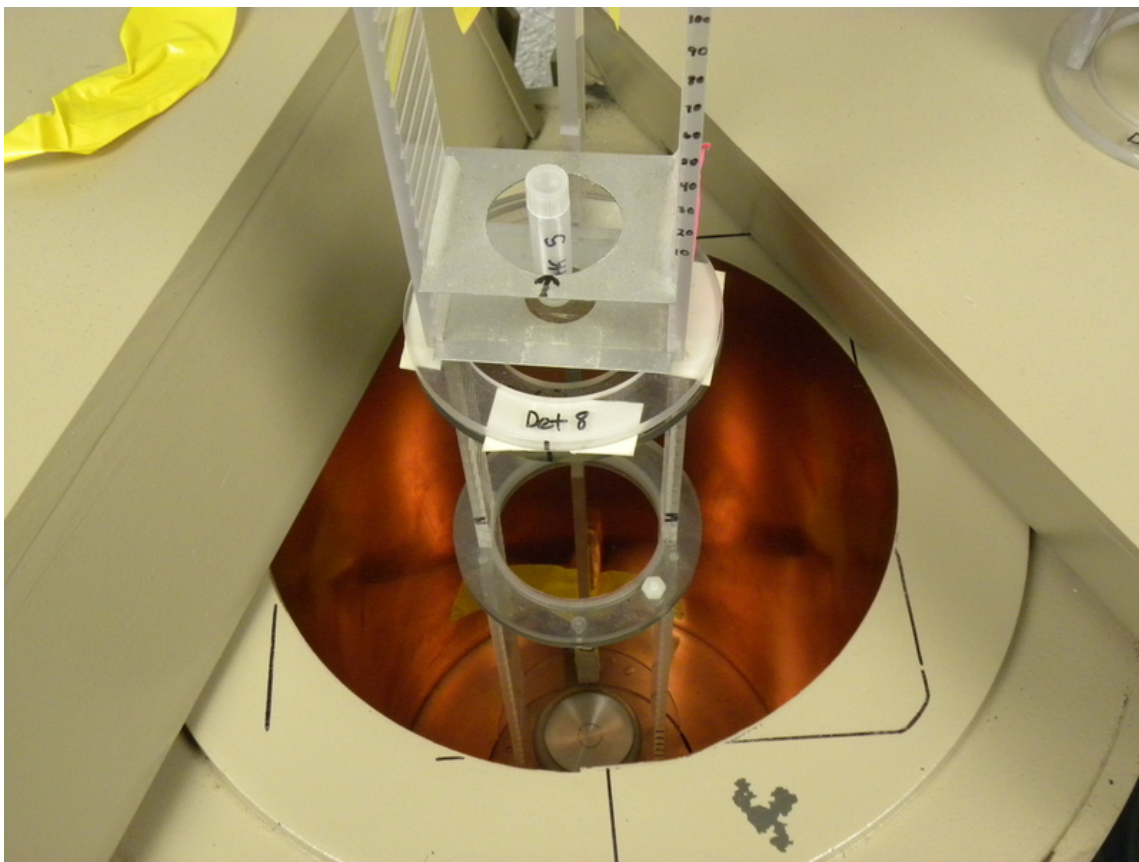


Figure 8.6: A counting vial is placed on the counting stand.

8.5 Solid Phase Taggants

The measured taggant vectors are shown in Table 8.7. The error of the prepared taggant vectors along with the error of the measured taggant vectors are shown in Table 8.8. The prepared error represents the statistical uncertainty associated with parameters that comprise the calculation of the prepared taggant vectors in Table 8.6. The measured error is the length of the vector formed between the measured taggant vectors in Table 8.7 and the prepared taggant vectors in Table 8.6.

The taggants are graphically represented in Figs. 8.7, 8.8, and 8.9.

Table 8.7: Measured taggants of solid phase powder samples (‰)

Solid Sample	¹⁰⁶ Cd	¹⁰⁸ Cd	¹¹⁴ Cd	¹²² Te	¹²⁴ Te	¹²⁶ Te	¹²⁸ Te
1	05224	00000	02716	00290	00000	01768	00000
2	05029	01250	00550	00453	00000	02716	00000

Table 8.8: Solid phase prepared and measured error

Taggant	Prepared error	Measured error
1	17.91	17.09
2	32.73	16.28

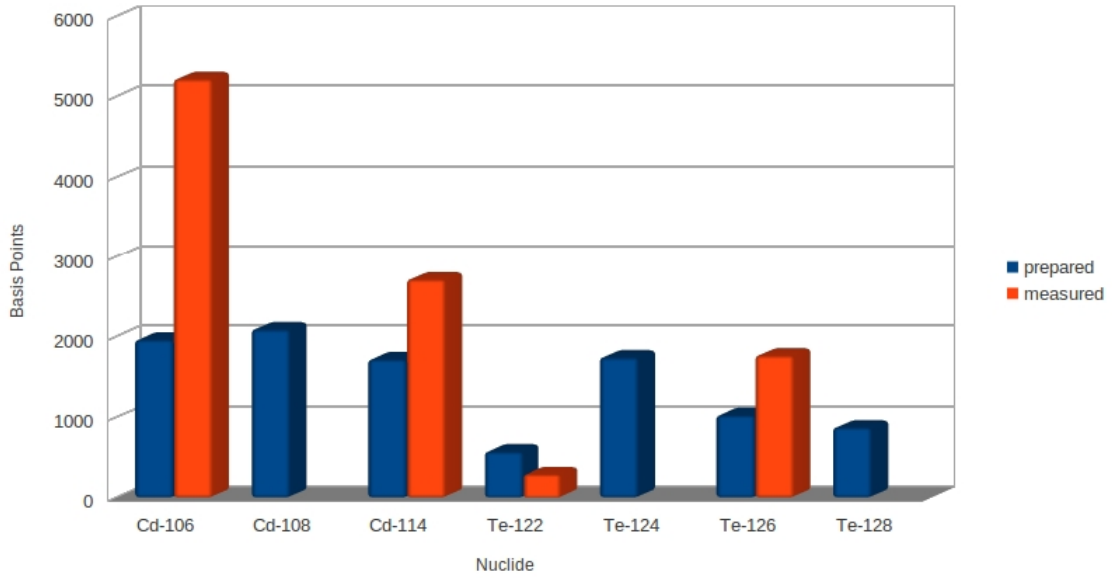


Figure 8.7: Measured and prepared taggant vectors of solid phase taggant 1.

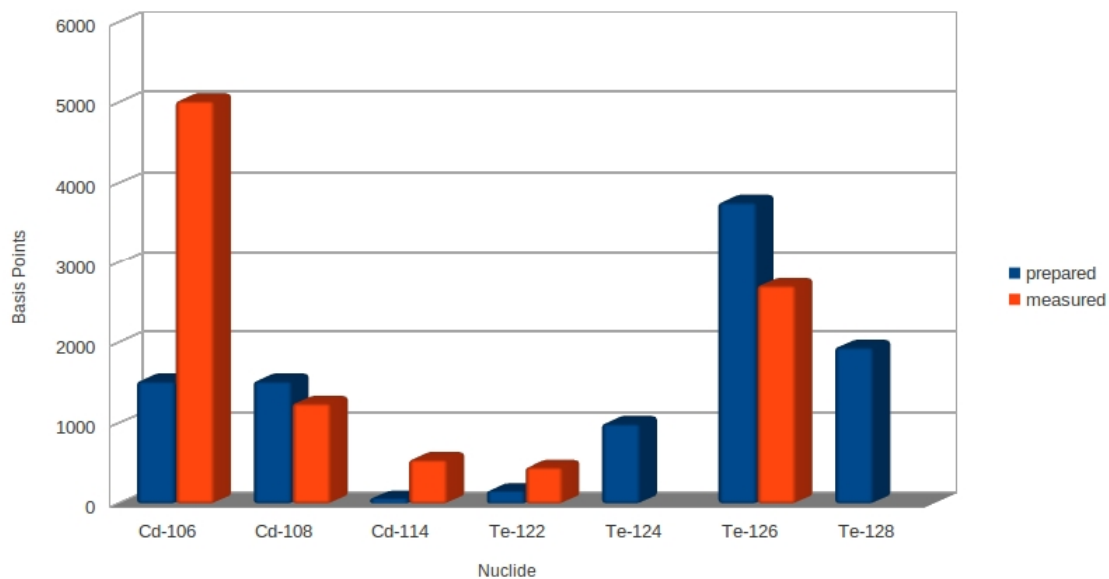


Figure 8.8: Measured and prepared taggant vectors of solid phase taggant 2.

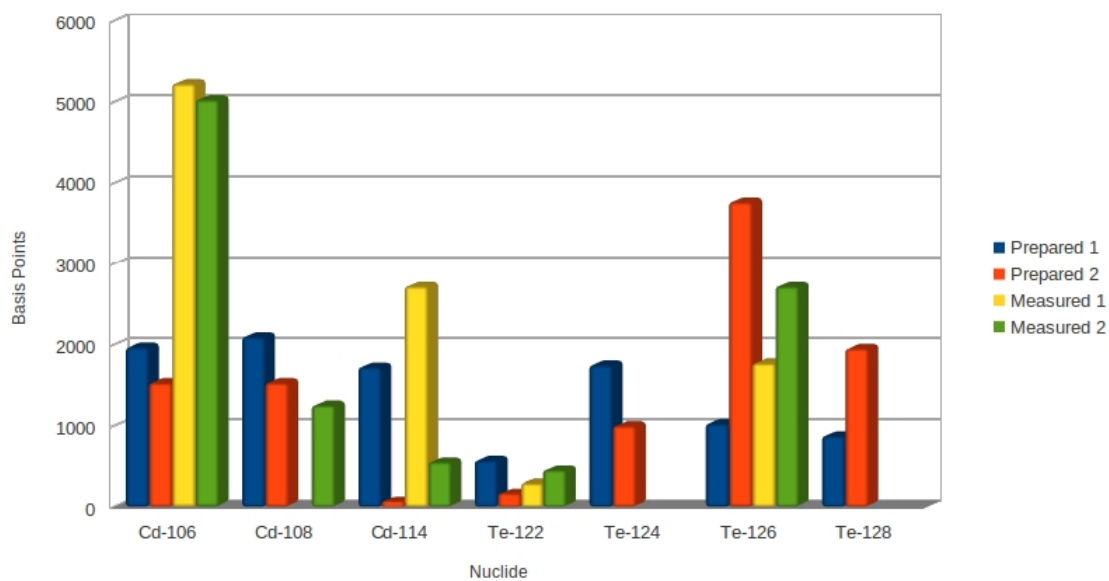


Figure 8.9: Measured and prepared taggant vectors of both solid phase taggants.

The differences shown in Fig. 8.9 indicate process loss during manufacture of the taggants after their initial mass was recorded. Errors in measurement of the taggants through NAA also contribute to this discrepancy. If there were no process loss during manufacture, the prepared and measured taggant values would be equivalent. While not equivalent, a connection can be observed in ^{108}Cd where the prepared and measured values of taggant 2 are both about 1000‰-1600‰. Observation of ^{114}Cd also is in agreement for taggant 2. Where equivalence is not present is most clearly shown in ^{106}Cd , ^{124}Te , and ^{128}Te . The prepared taggant values of ^{106}Cd both fall below 2000‰. The measured values of the same nuclide are at or above 5000‰. The absence of any measurement of ^{124}Te and ^{128}Te illustrates the discrepancy for those isotopes.

In addition to examining nuclides individually, an evaluation of all nuclides present in Fig. 8.9 as a whole assert their correlation to each other. Each nuclide is affected by shifts and anomalies in all other nuclides because of interdependence. If the measured values of ^{124}Te and ^{128}Te were instead in the range of 1000‰-2000‰, as expected, it may have reduced the difference between the prepared and measured value of ^{106}Cd to a more reasonable level. An error in measurement of one nuclide will affect the measured value of all other nuclides present in the taggant. The measured values do form a loose trend by following their corresponding prepared values, but are still vastly different at points, especially for ^{106}Cd in both taggants. Regardless of the presence of correlation between nuclides, the errors associated with the use of solid phase taggants was so great that improvement was needed before a functional taggant system was possible.

8.6 Regrouping

8.6.1 *Slippery When Dry*

Improvements were made to the process of taggant manufacture to reduce the observed error. Instead of using solid powders, a liquid transfer process was proposed. Residual dust from the powder was found remaining in shipping containers, plastic boats, and surfaces with which the batches came into contact. During pouring from one container to another, material is left behind. This occurs unevenly due to the different forms and fineness of the powder. The cadmium oxides, similar to grains of sand, rolled off the plastic boats with little residue while the fine tellurium metal powder left a fine dust in the plastic boats. The mass of this dust matters greatly when only a small amount of total material is involved, constituting a greater fraction of the small whole. This could also be an explanation as to why the measured cadmium values tended to outperform their prepared values while the measured tellurium values tended to underperform against their prepared values.

8.6.2 *Achieving a Particular Mass*

Measuring the difference between prepared and measured isotopic vectors does not account for the difficulty in preparing a certain desired vector. The desired optimal masses shown in Table 8.3 are vastly different than the prepared masses shown in Table 8.4. Pouring material manually was deemed to be an inefficient way to achieve a desired mass. Electrostatic force occasionally caused the tellurium metal to travel upwards along the plastic boat during the pouring process. This contributed to the error as the irradiated mass differed from the recorded mass due to electrostatic force after weighing.

8.6.3 *Change in Lineup*

The lineup of batches was changed to incorporate lessons and observations from the irradiation and counting process. The batches containing ^{108}Cd and ^{124}Te were dropped due to poor or no detection in the gamma-ray spectra. With only a gamma-ray at an energy of 109.29 keV with a 0.281% intensity, the $^{125\text{m}}\text{Te}$ did not produce enough activity for detection. The steep prices of the batches was also a factor in their dismissal.

Although the batch containing ^{128}Te never appeared in the taggant vector, shown in Table 8.7, it was found through NAA that the isotope of ^{128}Te constituted a 0.06% atomic fraction of the isotopic vector. This translated to less than the 0.01% cutoff for inclusion in the taggant vector. The low price allowed retention of the batch for further exploration.

In both samples, trace amounts of the isotope $^{131\text{m}}\text{Te}$ appeared from the activation of residual amounts of ^{130}Te present in the batches. With a low price, ^{130}Te was added to the lineup.

9. LIQUID PHASE

9.1 Initial Preparation of the Liquid Phase

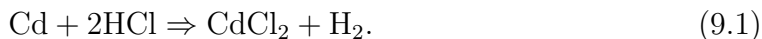
To ensure a higher degree of accuracy in measuring sample mass during the manufacture of each taggant, batches of stable isotopes were weighed in bulk before dissolution into the liquid phase. The optimal masses of each batch expected to produce 370 kBq after irradiation were modified from those in Table 8.3 incorporating experience gained through irradiations performed in Section 8. For new batches that were not previously irradiated, the computer code Isochain [45] was again used to determine optimal mass. These masses are shown in Table 9.1. Stable isotope batches were ordered in bulk to fulfill the material needs of 20 irradiations. This was deemed a sufficient sample size to adequately determine the efficacy of a taggant system without becoming prohibitively expensive.

Table 9.1: Batches used for liquid phase taggants

Batch	Primary Isotope	Optimal Mass (mg taggant ⁻¹)	Purchased Batch Mass (mg)
213001	¹⁰⁶ Cd	0.1	30.86
190842	¹¹⁴ Cd	0.1	8.13
206246	¹²² Te	0.2	2.4
187146	¹²⁶ Te	0.1	88.98
192241	¹²⁸ Te	4.0	59.87
187340	¹³⁰ Te	1.0	15.19

9.1.1 Dissolution in Acid

Two dissolutions were necessary as a significant volume of gas would be lost in the first reaction and an accurate concentration level could not be maintained with this loss. The cadmium was dissolved in OmniTrace®Ultra 37% HCl. The chemical reaction [48] is:



The tellurium was dissolved in OmniTrace®Ultra 70% HNO₃. After consulting several references [49, 50, 51] it appears that the exact mechanism for the dissolution of Te is not completely understood. The most likely scenario is that the dissolution of Te leads to the production of TeO₂, possibly followed by reaction with H₂O to form H₂TeO₃. After dissolution in acid, samples were evaporated to dryness.

9.1.2 Just Add Water

For the second and final dissolution, de-ionized water was prepared to a resistivity of 18 MΩ cm using a system by Nanopure™. This water was used to dissolve the CdCl₂ and HCl was used to dissolve the TeO₂. Information for the dissolved batches is shown in Table 9.2. The optimal volume contains enough batch material equivalent to the optimal mass calculated in Table 9.1.

9.1.3 Distribution to Irradiation Ampoules

The bulk volume of each batch was divided among 20 different irradiation ampoules by pipetting liquid volumes according to Table 9.3, distributing the masses shown in Table 9.4 into each irradiation ampoule. The concentrations are given in Table 9.2.

The prepared isotopic vectors of the taggants are shown in Table 9.5. Vectors

Table 9.2: Dissolved batch concentrations

Batch	Bulk Volume (mL)	Concentration ($\mu\text{g } \mu\text{L}^{-1}$)	Optimal Volume (μL)
^{106}Cd	12.344	2.5	40
^{114}Cd	4.878	1.66	60
^{122}Te	2.4	1	200
^{126}Te	17.796	5	20
^{128}Te	2.9935	20	200
^{130}Te	1.519	10	100

Table 9.3: Batch liquid volume in liquid phase taggants (μL)

Taggant	^{106}Cd	^{114}Cd	^{122}Te	^{126}Te	^{128}Te	^{130}Te
1	40	60	200	20	200	100
2	40	60	200	20	200	100
3	40	60	200	20	200	100
4	40	60	200	20	200	100
5	0	60	200	20	200	100
6	40	0	200	20	200	100
7	40	60	0	20	200	100
8	40	60	200	0	200	100
9	40	60	200	20	0	100
10	40	60	200	20	200	0
11	40	60	0	20	0	100
12	40	60	0	20	0	0
13	60	40	100	40	90	50
14	80	120	0	40	100	40
15	20	30	100	10	200	50
16	80	120	100	40	400	200
17	45	60	200	20	200	75
18	120	20	0	40	0	0
19	75	80	0	20	0	0
20	40	180	100	50	200	100

were randomly distributed near the optimal NAA isotopic vector and were designed to produce measurable differences when observed with NAA. Some taggants were also created by the absence of certain nuclides from the optimal isotopic vector.

Table 9.4: Batch mass in liquid phase taggants (μg)

Taggant	^{106}Cd	^{114}Cd	^{122}Te	^{126}Te	^{128}Te	^{130}Te
1	100	100	200	100	4000	1000
2	100	100	200	100	4000	1000
3	100	100	200	100	4000	1000
4	100	100	200	100	4000	1000
5	0	100	200	100	4000	1000
6	100	0	200	100	4000	1000
7	100	100	0	100	4000	1000
8	100	100	200	0	4000	1000
9	100	100	200	100	0	1000
10	100	100	200	100	4000	0
11	100	100	0	100	0	1000
12	100	100	0	100	0	0
13	150	66.6	100	200	1800	500
14	200	200	0	200	2000	400
15	50	50	100	50	4000	500
16	200	200	100	200	8000	2000
17	112.5	100	200	100	4000	750
18	300	33.3	0	200	0	0
19	187.5	133.3	0	100	0	0
20	100	300	100	250	4000	1000

This can be seen in Tables 9.3 and 9.4, where taggants 5-10 are simply the optimal isotopic vector with one nuclide absent.

After distribution, all liquid was allowed to evaporate to dryness to conform to the HFIR policy of preventing irradiation of liquid in sealed irradiation containers.

9.1.4 Sealing

The quartz ampoules were sealed as shown in Fig. 8.4, and placed into a graphite rabbit for irradiation. During the sealing evolution, taggant 11 was inadvertently crushed by tweezers immediately after sealing, taggant 6 cracked during the cooling process, and taggant 19 was not accepted by HFIR staff for irradiation due to

Table 9.5: Prepared taggant vectors of liquid phase taggants (‰)

Taggant	¹⁰⁶ Cd	¹¹⁴ Cd	¹²² Te	¹²⁶ Te	¹²⁸ Te	¹³⁰ Te
1-4	00188	00204	00380	00184	07257	01787
5	00000	00208	00387	00188	07396	01821
6	00192	00000	00388	00188	07408	01824
7	00196	00212	00000	00192	07543	01858
8	00192	00208	00387	00000	07393	01821
9	00686	00743	01385	00672	00000	06514
10	00229	00248	00463	00224	08836	00000
11	00796	00862	00000	00780	00000	07562
12	03266	03536	00000	03198	00000	00000
13	00550	00265	00370	00718	06359	01740
14	00685	00742	00000	00671	06602	01301
15	00109	00118	00220	00107	08410	01036
16	00194	00210	00098	00190	07470	01839
17	00222	00213	00397	00192	07577	01399
18	05640	00678	00000	03682	00000	00000
19	04373	03350	00000	02278	00000	00000
20	00179	00583	00181	00439	06915	01703

inadequate sealing.

9.2 Irradiation and Counting

Taggants were again irradiated in HFIR for a period of 1 hour and counted in a similar manner as described in Section 8. After irradiation, it was discovered that the quartz ampoule for taggant 18 had shattered during irradiation, but was contained within the graphite rabbit. During handling of the failed ampoule, a pair of outer gloves that had come into contact with the contaminated graphite rabbit were found to have a dose rate of $100 \mu\text{Sv h}^{-1}$ at 30 cm. Investigation of the gloves found the contamination to originate from ²⁴Na, a common activation product of aluminum.

9.3 Liquid Phase Taggants

9.3.1 Comparative NAA

After comparative NAA was performed on each sample, the Gharv program was used to determine the resulting isotopic vector and subsequent taggant vector. The measured taggant vectors are shown in Table 9.6. The prepared and measured taggant vector errors are shown in Table 9.7, which is also shown graphically in Fig. 9.1 to illustrate the stark difference between the prepared error and the measured error.

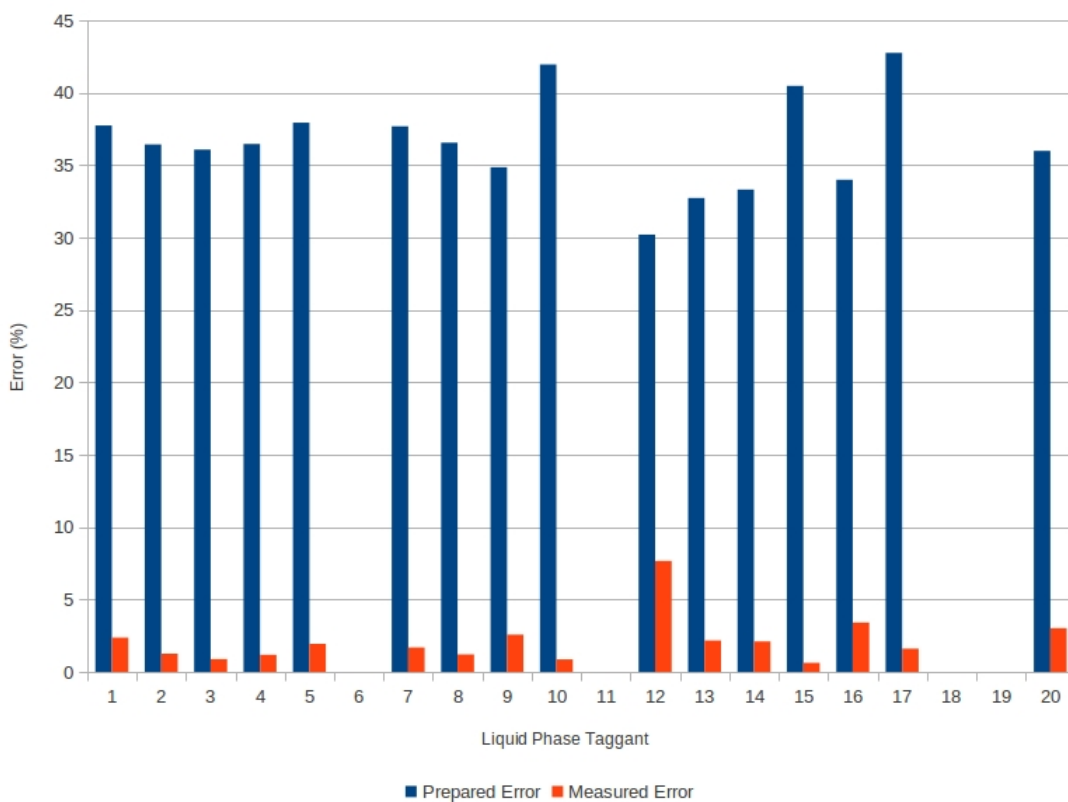


Figure 9.1: Prepared and measured error of liquid phase taggants.

Table 9.6: Measured isotopic vectors of liquid taggants (‰)

Taggant	¹⁰⁶ Cd	¹¹⁴ Cd	¹²² Te	¹²⁶ Te	¹²⁸ Te	¹³⁰ Te
1	00176	00128	00206	00050	07774	01664
2	00281	00199	00305	00082	07483	01647
3	00299	00195	00312	00090	07385	01716
4	00265	00187	00284	00087	07483	01690
5	00000	00181	00256	00071	07812	01677
6	-	-	-	-	-	-
7	00336	00222	00000	00093	07782	01565
8	00345	00215	00306	00000	07518	01614
9	01040	00751	01173	00351	00322	06361
10	00367	00250	00369	00102	08878	00031
11	-	-	-	-	-	-
12	04647	03378	00001	01934	00038	00000
13	00832	00249	00294	00341	06591	01690
14	00973	00676	00000	00310	06813	01226
15	00167	00120	00197	00049	08510	00954
16	00500	00347	00126	00128	06766	02130
17	00401	00242	00282	00081	07805	01185
18	-	-	-	-	-	-
19	-	-	-	-	-	-
20	00478	00902	00224	00365	06323	01705

Taggants 1-4 were manufactured to be identical to each other with the optimal mass for each nuclide and were used as the reference standard for comparative NAA. These four taggants yielded an average isotopic vector. This average vector was then calibrated to the manufactured isotopic vector, found in Table 9.5. All values in Tables 9.6 and 9.7 use this isotopic vector calibration standard as a reference point for the use of comparative NAA.

The large prepared error of each taggant is due to the multiple uncorrelated measurements that must be performed for each nuclide. Each nuclide used in the preparation of a taggant must be weighed individually, dissolved many times, and properly distributed to irradiation ampoules. There are many sources of possible

Table 9.7: Liquid phase prepared and measured error

Taggant	Prepared Error (%)	Measured Error (%)
1	37.74	2.37
2	36.43	1.26
3	36.07	0.89
4	36.46	1.19
5	37.93	1.94
6	-	-
7	37.68	1.69
8	36.55	1.21
9	34.85	2.58
10	41.95	0.87
11	-	-
12	30.20	7.67
13	32.73	2.17
14	33.31	2.11
15	40.47	0.63
16	33.99	3.41
17	42.75	1.61
18	-	-
19	-	-
20	35.98	3.02

measurement error at various points when this method is used and the errors are strictly additive. It is also important to note that the prepared error is an upper error bound, shown in Eq. 5.3. It is then expected that the actual prepared error will be lower than the values shown in Table 9.7.

The relatively lower measured error is most likely due to the advantage of correlation inherent in the measured error. The measured error is a collective, correlated assessment of the departure of the measured taggant vector from the prepared taggant vector. As mentioned in Section 8, the error in one nuclide is usually offset by an equal and opposite error in one or more other nuclides to maintain a normalized taggant vector length of unity. This property tends to limit the additive penalty of

errors in the correlated measured error but does nothing to prevent error accumulation in the uncorrelated prepared error. Taggantss 9 and 12 have a significantly lower measured error, but this is probably due to the reduced number of batches used to manufacture these taggants. Taggant 12 only consisted of 3 batches and only these 3 batches were detected during analysis, greatly reducing the error.

To reduce the prepared error upper bound, the experimental preparation methods could be refined to provide less error through process improvement. A rigorous sensitivity analysis can also be performed in place of using Eq. 5.3 to obtain an accurate prepared error estimate. If possible, repeated experiments for each isotopic vector will yield an accurate measured error, which can be used in place of a calculated prepared error, but will most likely be prohibitively expensive.

9.3.2 Parametric NAA

If parametric NAA is performed instead of comparative NAA, the isotopic vectors and associated errors are found in Tables 9.8 and 9.9.

The parametric NAA prepared error in Table 9.9 is lower than the comparative NAA prepared error in Table 9.7. This is unexpected as parametric NAA uses more input parameters in formation of the prepared taggant vector and should, in theory, have higher error. The measured error of parametric NAA is higher than comparative NAA, as expected, which further indicate that the uncorrelated methods used in formation of the prepared taggant vector may need improvement.

9.3.3 Uncorrelated Taggants

While correlated error is shown in Table 9.7, the uncorrelated error of each individual nuclide associated with comparative NAA is shown in Table 9.10. Uncorrelated measurements for the solid phase taggants are shown in Fig. 8.9.

Similar to Section 8, there are two nuclides which have clear trends across all

Table 9.8: Measured taggant vectors of liquid taggants using parametric NAA (‰)

Taggant	¹⁰⁶ Cd	¹¹⁴ Cd	¹²² Te	¹²⁶ Te	¹²⁸ Te	¹³⁰ Te
1	00897	01136	00436	00999	00600	05929
2	01221	01510	00549	01241	00484	04993
3	01256	01432	00543	01295	00457	05014
4	01154	01421	00513	01300	00483	05125
5	00000	01557	00543	01312	00600	05986
6	-	-	-	-	-	-
7	01498	01729	00000	01358	00529	04883
8	01627	01781	00600	00121	00542	05327
9	01277	01607	00596	01273	00000	05243
10	02411	02870	01006	02251	00970	00490
11	-	-	-	-	-	-
12	02936	03717	00000	03346	00000	00000
13	02274	01242	00333	02707	00243	03198
14	02430	02944	00000	02229	00247	02147
15	01130	01414	00554	01308	00920	04671
16	01602	01939	00167	01287	00304	04699
17	01832	01945	00535	01286	00552	03847
18	-	-	-	-	-	-
19	-	-	-	-	-	-
20	01081	03477	00209	02376	00190	02664

taggants. The clear positive bias of ¹⁰⁶Cd is offset by a clear negative bias of ¹²⁶Te. This trend shows the clear advantage of using correlated measured taggant vectors because an error in any one nuclide will tend to affect the combined correlated error less than the combined uncorrelated error, resulting in a lower correlated measured error. The uncorrelated error of individual nuclides is not without utility though. Systemic bias can be identified and corrected for, such as ¹⁰⁶Cd and ¹²⁶Te in Table 9.10. Uncorrelated error can also be used if known elemental fractionation, the selective chemical separation in a compound, occurs or if a known isotope has been added or removed from the system. This can be used to correct for known spoofing or to identify specific anomalies that would be missed with only correlated error.

Table 9.9: Liquid phase prepared and measured error using parametric NAA

Taggant	Prepared Error	Measured Error
1	29.07	32.53
2	25.07	31.64
3	25.21	31.77
4	25.55	31.81
5	29.35	33.33
6	-	-
7	24.80	32.59
8	26.25	32.62
9	26.36	7.85
10	13.66	36.09
11	-	-
12	17.22	1.65
13	20.07	28.11
14	16.35	29.27
15	23.83	35.02
16	24.00	33.09
17	20.78	32.18
18	-	-
19	-	-
20	18.03	31.38

Table 9.10: Uncorrelated difference between prepared and measured taggant vectors of liquid phase taggants (%)

Taggant	^{106}Cd	^{114}Cd	^{122}Te	^{126}Te	^{128}Te	^{130}Te
1	-6.38	-37.25	-45.79	-72.83	7.12	-6.88
2	49.47	-2.45	-19.74	-55.43	3.11	-7.83
3	59.04	-4.41	-17.89	-51.09	1.76	-3.97
4	40.96	-8.33	-25.26	-52.72	3.11	-5.43
5	-	-12.98	-33.85	-62.23	5.62	-7.91
6	-	-	-	-	-	-
7	71.43	4.72	-	-51.56	3.17	-15.77
8	79.69	3.37	-20.93	-	1.69	-11.37
9	51.60	1.08	-15.31	-47.77	-	-2.35
10	60.26	0.81	-20.30	-54.46	0.48	-
11	-	-	-	-	-	-
12	42.28	-4.47	-	-39.52	-	-
13	51.27	-6.04	-20.54	-52.51	3.65	-2.87
14	42.04	-8.89	-	-53.80	3.20	-5.76
15	53.21	1.69	-10.45	-54.21	1.19	-7.92
16	157.73	65.24	28.57	-32.63	-9.42	15.82
17	80.63	13.62	-28.97	-57.81	3.01	-15.30
18	-	-	-	-	-	-
19	-	-	-	-	-	-
20	167.04	54.72	23.76	-16.86	-8.56	0.12

10. COMMON RESEARCH REACTORS

10.1 Portability

One of the factors affecting the use of a taggant system is the relative ease of use in obtaining information about the taggant. This can be an advantage or disadvantage depending on the situation. A system that requires many steps to be completed will make identification of the taggant more difficult but will also prevent unwanted detection of the taggant by third parties. To test the portability of the taggant, the Texas A&M University 1 MW_{th} TRIGA reactor was employed. A positive outcome using a smaller reactor would allow greater portability of the taggant, but would also open up the possibility of nefarious uses and modification of the taggant.

10.2 Preparation

10.2.1 Dissolution

A small sample size was manufactured with only 4 taggants. Using the liquid phase methods described in Section 9, each taggant was manufactured to be identical to the other taggants, mimicking the motivation behind the manufacture of taggants 1-4 in Table 9.5. In addition, an empty irradiation vial was filled with deionized water and evaporated to dryness to test for impurities in the de-ionized water that may affect the outcome. It was found that all impurities of cadmium and tellurium in the water and vial were below minimum detectable levels. The final dissolution information is shown in Tables 10.1 and 10.2. Masses were obtained using a Mettler Toledo XP105 delta range balance with 10 μ g resolution.

The taggant vectors are shown in Table 10.3. These vectors are different than the previous vectors in Table 9.5 because of the differences in allowed irradiation time,

Table 10.1: Dissolved batch concentrations for a common research reactor

Batch	Bulk Volume (mL)	Concentration ($\mu\text{g } \mu\text{L}^{-1}$)	Optimal Volume (μL)
^{106}Cd	3.2	1.1750	800
^{114}Cd	4.0	2.2550	1000
^{122}Te	1.2	1.5250	300
^{126}Te	0.8	1.0500	200
^{128}Te	0.4	1.1500	100
^{130}Te	0.2	1.0000	50

Table 10.2: Liquid volume and mass of common research reactor taggants

Isotope	Volume (μL)	Mass (μg)
^{106}Cd	800	940
^{114}Cd	1000	2255
^{122}Te	300	457.5
^{126}Te	200	210
^{128}Te	100	115
^{130}Te	50	50

decay time, and reduction in neutron fluence rate afforded by the smaller TRIGA reactor.

Table 10.3: Prepared taggant vectors of common research reactor taggants (‰)

Sample	^{106}Cd	^{114}Cd	^{122}Te	^{126}Te	^{128}Te	^{130}Te
1-4	02234	05803	01097	00488	00263	00112

10.2.2 Polyethylene Irradiation Containers

Instead of using a quartz ampoule inside of a graphite rabbit as previously used in Sections 8 and 9, the irradiation vials were made of polyethylene. The vials were

manufactured by LA Container (Yorba Linda, CA) and two small vials were placed inside of a large vial before irradiation to prevent failure. After the second bulk dilution of the stable isotope batches, the remaining liquid of each taggant was pipetted into two small vials, model EP338NAA, with an inner diameter of 10 mm, height of 22 mm, and a maximum volume of 1.40 mL. Each small vial contained approximately half of the liquid volume of each taggant. The vials were then evaporated to dryness and sealed with a soldering iron. The small vials were then placed inside of a large vial, model EP290NAA, measuring 14 mm inner diameter, 54 mm height, and a maximum volume of 8.06 mL. The large vial was then sealed in a similar manner, creating two air-tight barriers around the taggant.

10.3 Irradiation

Taggants were irradiated for 16 minutes in the Texas A&M TRIGA reactor pneumatic system, position D2, at a power of 1 MW_{th} . While previous irradiations in Sections 8 and 9 took place for 1 hour, the irradiation time was reduced to 16 minutes due to differences in the pneumatic sample container. The polyethylene vials used for irradiation in the TRIGA reactor cannot withstand a high neutron fluence and are limited to 16 minute irradiations to prevent sample failure.

10.4 Counting

The counting of the samples was allowed to commence much quicker after irradiation in the Texas A&M counting laboratory. The procedures for extraction from the reactor, along with health physics precautions, allowed samples to be counted within 5 minutes after removal from the reactor core. Samples were counted on a HPGe for 1 hour at a vertical distance of 90 cm. For the activation of ^{128}Te and ^{130}Te , focus was shifted from the ground states of ^{129}Te and ^{131}Te after activation to the short lived $^{129\text{m}}\text{Te}$ and $^{131\text{m}}\text{Te}$ metastable states, allowing for a reduction in initial mass

for irradiation. This was possible because the counting of samples was allowed to commence much quicker after irradiation in the Texas A&M counting laboratory. By using a smaller reactor and reducing the decay time, only the taggant vector needed to be changed to accommodate the reduction in neutron fluence.

10.4.1 Comparative NAA

The measured taggant vectors are shown in Table 10.4. The error of the taggant vectors is shown in Table 10.5. To complete the comparative NAA shown in Tables 10.4 and 10.5, the average activity of each nuclide for all taggants was found and all taggants were compared to this average.

Table 10.4: Measured taggant vectors of common research reactor taggants using comparative NAA (‰)

Taggant	¹⁰⁶ Cd	¹¹⁴ Cd	¹²² Te	¹²⁶ Te	¹²⁸ Te	¹³⁰ Te
1	03212	04976	01086	00422	00240	00061
2	02958	05343	01034	00365	00210	00087
3	00906	06637	01428	00639	00273	00114
4	03422	04812	01053	00424	00199	00087

Table 10.5: Common research reactor prepared and measured error using comparative NAA

Taggant	Prepared Error	Measured Error
1	34.87	5.24
2	32.26	3.56
3	17.04	6.57
4	37.17	6.33

As expected, the prepared error of the taggants were similar to the prepared error in Table 9.7. This is expected as the liquid phase preparation method used in Section 9 was followed as close as practicable. It is also expected that the measured error is far below the prepared error but still slightly higher than the prepared error in Table 9.7. As mentioned in Section 7, the HFIR neutron fluence rate is one of the highest in the world and certainly higher than a moderate university research reactor. This increased rate combined with longer irradiation time allowed greater activation to take place in the HFIR, which decreased the overall error when gamma-ray spectroscopy was performed.

10.4.2 Parametric NAA

Parametric NAA was also employed and the measured taggant vectors are shown in Table 10.6 with the corresponding error shown in Table 10.7. It should be noted in Tables 10.5 and 10.7 that taggants 1 and 4 experienced a decrease in error, while taggant 2 experienced a slight increase in error. Taggant 3 underwent the largest error increase of all when parametric NAA was used. By averaging the activities, the error is more distributed across all of the taggants.

Table 10.6: Measured taggant vectors of common research reactor taggants using parametric NAA (‰)

Taggant	¹⁰⁶ Cd	¹¹⁴ Cd	¹²² Te	¹²⁶ Te	¹²⁸ Te	¹³⁰ Te
1	02969	05748	00331	00532	00358	00058
2	02713	06118	00313	00458	00314	00081
3	00819	07457	00426	00786	00403	00105
4	03174	05582	00322	00536	00301	00082

Unlike the parametric NAA trends in Section 9, both the prepared error and

Table 10.7: Common research reactor taggant vector error using parametric NAA

Taggant	Prepared Error	Measured Error
1	63.83	4.37
2	67.01	3.98
3	79.33	9.39
4	62.59	5.06

measured error, on average, increased when parametric NAA was used. The increase of the prepared error is sharp, roughly by a factor of 2 for all samples. The rise in the measured error average is mainly due to the increase in error of taggant 3, indicating an anomaly with the taggant.

10.4.3 Uncorrelated Taggants

The uncorrelated error using comparative NAA is shown in Table 10.8.

Table 10.8: Uncorrelated difference between prepared and measured taggant vectors of taggants used in common research reactors (‰)

Taggant	¹⁰⁶ Cd	¹¹⁴ Cd	¹²² Te	¹²⁶ Te	¹²⁸ Te	¹³⁰ Te
1	43.78	-14.25	-1.00	-13.52	-8.75	-45.54
2	32.41	-7.93	-5.74	-25.20	-20.15	-22.32
3	-59.44	14.37	30.17	30.94	3.80	1.79
4	53.18	-17.08	-4.01	-13.11	-24.33	-22.32

Similar to Section 9, ¹⁰⁶Cd had a clear positive trend, but unlike Section 9, the negative errors were distributed among the other nuclides instead of one specific nuclide. As with Table 10.7, taggant 3 was an outlier with ¹⁰⁶Cd containing a negative error instead of a positive error. The uncorrelated errors were similar to those found in Table 9.10, affirming the utility of uncorrelated error analysis only for specific

situations and not for overall use.

11. SUMMARY

11.1 Assessment of the Taggant System

It was determined that a taggant system utilizing NAA is feasible if certain key problems are solved. While no taggant system will work for all applications, the solution of these problems will make the system generally attractive to a greater number of potential users.

11.1.1 Positive Aspects

11.1.1.1 Liquid Phase Preparation

The use of liquid phase preparation techniques in the manufacture of a taggant system has shown to be effective. Even when taggants were manufactured with little to no automation, the errors observed were acceptable for a robust taggant system. It is postulated that automation in the liquid phase manufacturing process would reduce errors even further.

11.1.1.2 NAA

NAA has proven an effective alternative to MS for isotopic analysis of the taggants. The lower precision of NAA compared to MS is compensated for by the rapid analysis capability of the NAA method. This is realized in Section 10 because a sample can be prepared, irradiated, and counted in a matter of a few hours.

The use of comparative NAA in Sections 9 and 10 has shown that it is more effective than parametric NAA in the attribution of specific taggants. A method to emphasize the accurate manufacture of a taggant reference standard would only serve to increase the accuracy of comparative NAA even more when used in the context of a taggant system. Parametric NAA would still have utility by itself or as

a preliminary guide in the selection of a reference standard for comparative NAA.

11.1.1.3 Low Mass Required

The low masses required of the stable batch isotopes were sufficiently small to allow concealment of the stable batch isotopes among objects to avoid detection. The reliable and consistent detection of all radionuclides by NAA affirmed that masses on the order of μg to mg were sufficient for a taggant system.

11.1.1.4 The Use of Common Research Reactors

If the isotopic vectors are adjusted, Section 10 shows promise for the use of taggants in common research reactors around the world. The effectiveness of using small reactors was similar to that of the much larger research reactors. For taggants not requiring the convenience of measurement around the world, isotopic vectors similar to those in Section 9 were useful in limiting the use of NAA to a small group of reactors.

11.1.2 Negative Aspects

11.1.2.1 Solid Phase Preparation

The process used in the manufacture and cataloging of prepared taggants is crucial to an accurate taggant system. Preparation of taggants without dilution to the liquid phase was wrought with problems in Section 8. These issues can be overcome either by using liquid phase preparation methods or through the manufacture of solid phase taggants in bulk quantities. By mixing large quantities of material at one time, the process loss of small amounts will not greatly affect the error of the prepared taggants.

11.1.2.2 Individually Manufactured Taggants

Liquid phase preparation would also benefit through the use of bulk manufacturing practices. The large throughput of a large volume of liquid would minimize the effect of the loss of 1 μg or 1 mg of material. Automation and uniformity of the distribution system would reduce errors associated with measurement and transfer of material during taggant manufacture.

11.1.2.3 Parametric NAA

In Sections 8, 9, and 10, parametric NAA fell short of comparative NAA in almost every way, as was expected. The error introduced by uncertainty in nuclear data and uncertainty about the neutron environment will always put parametric NAA at a disadvantage. If there are no reference standards, parametric NAA may be the only resource to obtain isotopic vectors, but it should be used only as a last resort.

11.1.2.4 Uncorrelated Measurements

Measurement of the change in individual nuclides showed little utility for use in a taggant system, as shown in Tables 9.10 and 10.8. In most cases, the error of one nuclide alone was greater than the combined Euclidean norm of all nuclides measured as a whole. The use of uncorrelated measurements will find application in the detection of anomalies, elemental fractionation, and detailed error analysis of individual taggants. While only correlated measurements can be used in the routine attribution of taggants to maintain acceptable error, uncorrelated measurements allow different questions to be answered. The elemental fractionation may provide information pertaining to a chemical process that was used on the taggant, such as the length of time since chemical purification if that taggant was placed in a radiological

material. If the taggant was exposed to a neutron field, such as in a nuclear reactor, the selective depletion of certain nuclides could yield information about the reactor if the original identity of the taggant was known. The uncorrelated measurements of nuclides may be just as useful or even more useful than correlated measurements depending on the nature of the information sought.

11.2 Cost

A taggant system is only useful if it is economically feasible for an organization to use it. The total cost per taggant is summarized in Tables 11.1, 11.2, and 11.3. The measured masses of the samples that were designated as the optimal masses were used for calculation to serve as a fair comparison between different phases and reactors.

As expected, the solid phase irradiations performed in HFIR were the most costly and least efficient. Prices are based on the measured balance mass, which differed greatly from the desired mass for the solid phase batches. If it were possible to pour exactly the desired amount onto the balance, the solid phase prices would more closely match that of the liquid phase. The prices take into account process loss, as the prices shown assume no material is lost. The masses stated are likely higher than the irradiated mass, but process loss must be taken into account.

In the liquid phase, it is more expensive to irradiate in a small common research reactor than a large reactor. While the small research reactor had the advantage of less decay time, the large research reactor had the dominating advantage of greater overall neutron fluence, which more than compensated for the greater decay time and allowed for the least expensive irradiation.

Table 11.1: Prices of taggants irradiated in solid phase

Batch	Unit Cost (\$ mg ⁻¹)	Mass (mg taggant ⁻¹)	Taggant Cost (\$ taggant ⁻¹)
Taggant 1			
¹⁰⁶ Cd	76.68	0.69	52.91
¹⁰⁸ Cd	100.81	0.75	75.61
¹¹⁴ Cd	2.09	0.64	1.34
¹²² Te	78.92	0.20	15.78
¹²⁴ Te	31.94	0.62	19.80
¹²⁶ Te	9.58	0.37	3.54
¹²⁸ Te	4.71	0.32	1.51
Total			170.49
Taggant 2			
¹⁰⁶ Cd	76.68	1.03	78.98
¹⁰⁸ Cd	100.81	1.05	105.85
¹¹⁴ Cd	2.09	0.05	0.10
¹²² Te	78.92	0.11	8.68
¹²⁴ Te	31.94	0.68	21.72
¹²⁶ Te	9.58	2.61	25.00
¹²⁸ Te	4.71	1.37	6.45
Total			246.78

Table 11.2: Prices of taggants irradiated in liquid phase

Batch	Unit Cost (\$ mg ⁻¹)	Mass (mg taggant ⁻¹)	Taggant Cost (\$ taggant ⁻¹)
¹⁰⁶ Cd	76.68	0.1	7.67
¹¹⁴ Cd	2.09	0.1	0.21
¹²² Te	78.92	0.2	15.78
¹²⁶ Te	9.58	0.1	0.96
¹²⁸ Te	4.71	4.0	18.84
¹³⁰ Te	4.81	1.0	4.81
Total			48.27

Table 11.3: Prices of taggants irradiated in a common research reactor

Batch	Unit Cost (\$ mg ⁻¹)	Mass (mg taggant ⁻¹)	Taggant Cost (\$ taggant ⁻¹)
¹⁰⁶ Cd	76.68	0.94	72.08
¹¹⁴ Cd	2.09	2.255	4.71
¹²² Te	78.92	0.4575	36.11
¹²⁶ Te	9.58	0.21	2.01
¹²⁸ Te	4.71	0.115	0.54
¹³⁰ Te	4.81	0.05	0.24
Total			115.69

11.3 Unique Identifiers

The number of possible unique identifiers is important in a regulated and diverse taggant system. A small number of unique taggants are adequate for simple tagging purposes, but a large number of unique taggants allows for extra flexibility and complexity.

11.3.1 Computational Method to Determine Number of Unique Taggants

A computer program was written to determine the number of possible combinations of taggants given certain enrichment increments. The program begins by setting all isotopes to 0% and incrementing the last isotope in the list by one enrichment increment for that particular isotope. If the sum of the combination is equal to 100%, the combination is recorded and tallied. If the sum of the combination is less than 100%, the last isotope in the list is continuously incremented until it meets or exceeds a sum of 100%. After the sum reaches 100%, the last isotope is reset to 0% and the penultimate isotope is incremented by one enrichment increment. This process is repeated until the first isotope in the list meets or exceeds 100% itself. By numerically incrementing each isotope, a complete list of unique taggants is created.

11.3.2 Enrichment Increments

Along with the Euclidean distance between the measured and prepared taggant that is the output of the Gharv code, a list of statistical errors by nuclide are included in the output. As the errors are correlated, there is no way to attribute statistical error to any one nuclide, so it is assumed that errors were distributed evenly across all nuclides and the resulting magnitudes of the individual errors were adjusted until the taggant vector matched the Euclidean distance. After this adjustment, errors were averaged across all measured taggants in a particular category and then rounded, usually up, to integers. To fulfill the requirement to sum to exactly 100%, it was desirable that each increment was a factor of as many other increments as possible to increase the number of possible combinations while still correctly approximating the correct error of each nuclide. If a nuclide was not detected, such as for ^{124}Te and ^{126}Te in the solid phase, it was arbitrarily assigned an enrichment increment of 1%. The errors of each nuclide are shown in Tables 11.4, 11.5, and 11.6.

Table 11.4: Enrichment increments of solid phase taggants

Nuclide	Individual Error (%)	Enrichment Increment (%)
^{106}Cd	57.06	60
^{108}Cd	9.185	10
^{114}Cd	14.5	15
^{122}Te	4.445	5
^{124}Te	0	1
^{126}Te	26.765	30
^{128}Te	0	1

Table 11.5: Enrichment increments of liquid phase taggants

Nuclide	Individual Error (%)	Enrichment Increment (%)
^{106}Cd	8.41	10
^{114}Cd	6.29	8
^{122}Te	3.29	4
^{126}Te	3.15	4
^{128}Te	77.9	80
^{130}Te	20.32	20

Table 11.6: Enrichment increments of common research reactor taggants

Nuclide	Individual Error (%)	Enrichment Increment (%)
^{106}Cd	35.08	35
^{114}Cd	62.84	60
^{122}Te	13.28	15
^{126}Te	5.23	5
^{128}Te	2.69	2
^{130}Te	1	1

11.3.3 Unique Combinations

The number of possible combinations of taggants using the enrichment increments of Tables 11.4, 11.5, and 11.6 are shown in Table 11.7.

Table 11.7: Unique number of possible taggants by category

Category	Number of Unique Taggants
Solid Phase	13965
Liquid Phase	864
Common Research Reactor	2257

The solid phase taggants most likely have such a high number of possible com-

binations because of the benefit of one additional nuclide. While the liquid phase taggants irradiated in the HFIR and TRIGA reactors are substantially similar and contain the same number of nuclides, they contain different optimal isotopic vectors. This difference may have contributed to the increased error associated with the optimal isotopic vector for HFIR.

11.3.4 Tailored Taggants

The differences in optimal isotopic vectors irradiated in the HFIR and the TRIGA illustrate the possibility of tailoring taggant vectors to different nuclear reactors for NAA purposes. A large reactor such as HFIR will provide the ability to irradiate and detect nuclides with a small mass or cross-section. Taggant vectors containing a large fraction of these nuclides will tend to be detectable only in large reactors such as HFIR, which require a significant capital investment to construct and operate. Conversely, taggants can be manufactured with taggant vectors that are suitable for smaller reactors with a rated power of less than $10 \text{ MW}_{\text{th}}$.

11.4 Spoofing

The integrity of a taggant system is paramount to security and reliability. No taggant system is foolproof, but there are certain features of the taggant system that make it resistant to spoofing and render it less likely to be compromised.

11.4.1 Artificial Material

The taggants are composed of artificially enriched isotopes that are not found in nature. It is not trivial to obtain these isotopes and combine them to form the pattern desired to form a particular taggant vector. This would have to take place after additional measurement of the taggant, which will also introduce error. The error associated with measurement of the taggant combined with the error in creation

of the spoofed taggant would most likely be far off the intended target and would not form a known taggant. The presence of artificially enriched material that does not form a known taggant would not necessarily prove that spoofing occurred but would certainly raise suspicion.

11.4.2 Hidden Taggants

An advantage of taggants made from stable isotopes at the atomic level is that they are so tiny that they are not visible. They will not be stumbled upon accidentally and are very hard to detect, unless it is known exactly where to look. This aspect adds to the taggant security by remaining unseen.

11.4.3 The Path to Natural Material

Adding artificially enriched material to the taggant would most likely create a bogus taggant and would indicate spoofing but not readily identify the original taggant. If it was known that only natural material was used to spoof the taggant, the resulting taggant vector would lie somewhere in the phase space between natural material and the original taggant. By tracing a vector originating at the natural material vector through the bogus taggant vector, the resulting vector would identify a small number of taggant vectors or even a single taggant vector that would be good candidates for the original taggant vector before spoofing. In this case, it would not only be known that the material was spoofed, but the original taggant vector would be known.

11.5 Future Work

11.5.1 Increasing the Number of Unique Combinations

It is always desirable to have the largest number of unique combinations in any tagging system. This is the result of a system in which there is little to no error

associated with each taggant and there are many components, or in this case nuclides, that form the basis of the taggant vector. By increasing the number of nuclides and reducing the error, each taggant vector will become more unique and harder to spoof. The option of using millions or billions of unique combinations will make a taggant system more attractive to potential users with more applications for use.

11.5.2 Cost

An taggant system that is vastly more expensive than the object that it is designed to protect may not be economically feasible for potential users. By streamlining the manufacturing process and reducing cost, the tagging system would become beneficial to more applications. Ways to drive the cost down would also benefit other aspects of the taggant system. A process that would use less material would reduce the initial cost of purchasing the material, but would also create a taggant that is smaller and harder to detect by prying eyes.

11.5.3 Attribution Confidence

No taggant system will always correctly attribute each taggant vector to the correct taggant. It would be beneficial to know the relative reliability of each taggant and the confidence associated with such a taggant. Some applications will demand a taggant with supreme confidence in areas of importance while other applications may not require such a high threshold.

11.5.4 Spoofing

Ways to detect if the taggant has been altered and by how much are always useful to detect and track nefarious purposes and motives. All taggants will most likely have a group or individual that is interested in compromising or spoofing the taggant. By implementing processes or safeguards to prevent or detect spoofing, the

integrity of the taggant system will be preserved. There will always be a way to spoof the system, but making it difficult will remove any possible benefits associated with spoofing.

11.5.5 Use of MS

The use of NAA is one in which the resulting isotopic vectors can be ascertained in a relatively short period of time. The accuracy and speed of NAA was once vastly superior to that of MS and led to widespread use of NAA in the last half of the 20th century. Advances in MS have decreased or removed the advantage that NAA once had over MS, making MS much more precise, yet still slower overall as a process when compared to NAA. Future advances and development of MS will almost certainly lead to more precise, quicker, and cheaper measurements, and could possibly make NAA obsolete. For certain applications that do not require a short response time but do require high precision, MS may already be the preferred method to analyze taggants instead of NAA. Any method capable of measuring isotopic vectors will be suitable in a taggant system as long as it is suitable for the application at hand in other areas such as cost and time of processing.

11.5.6 Minimum Detectable Mass

The use of a small amount of mass is desirable to avoid taggant detection by unwanted parties. The minimum detectable mass is determined by which particular isotopic vector is present and which method is used to form the isotopic vector. NAA will have a greater minimum detectable mass, generally in the μg -mg range while MS is able to detect much lower concentrations.

11.5.7 Different Elements

Cadmium and tellurium are only one choice of elements. There will be other elements better suited for particular applications and each application may be open to more than one element. The chemical and physical form of these elements will be based on the application and are not limited to nanoparticles, solid, or even liquid phase. Any element that can be isotopically enriched and combined with other elements or isotopes is a candidate for use in a taggant system.

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APPENDIX A

GHARV SOURCE CODE

Logic Flow

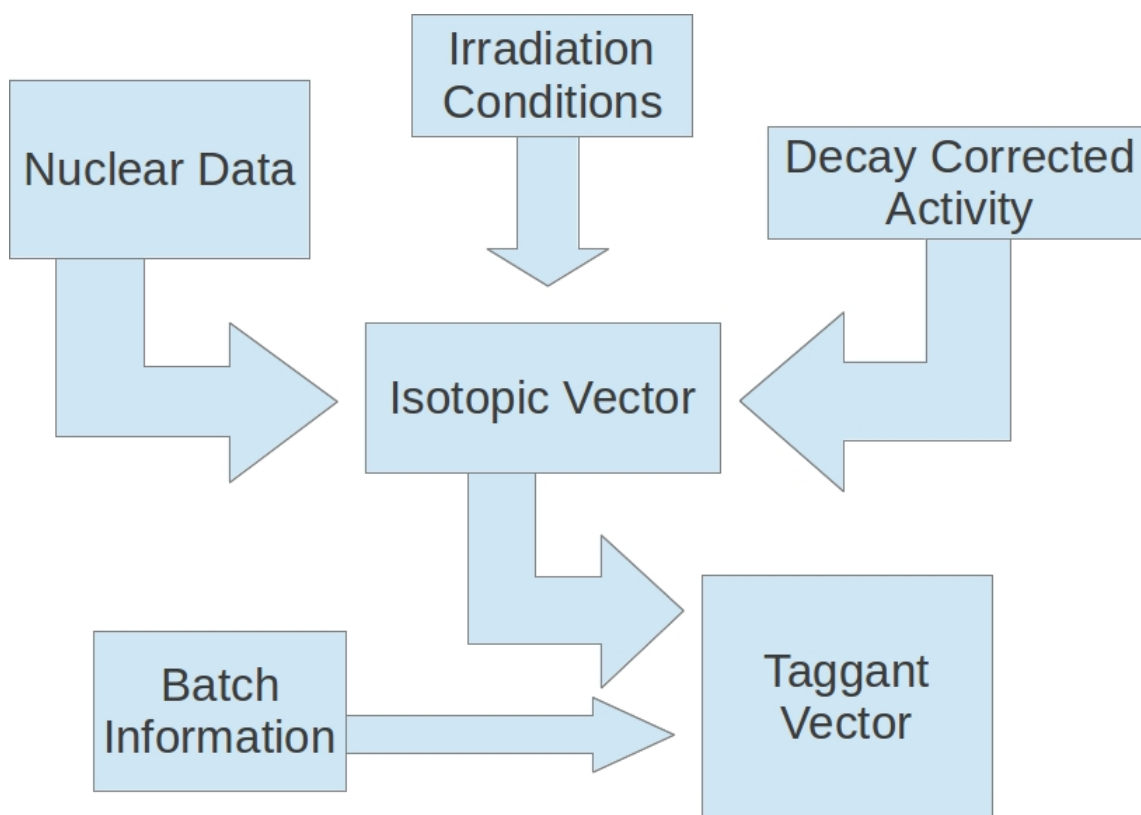


Figure A.1: Flowchart depicting the logic flow of the Gharv program.

Source Code

```
!this subroutine solves the AX=B problem and associated  
!error analysis to backcalcate what the barcode will be
```



```

!given the inputs of atomic ratios collected by NAA and
!mass spec assay data about batches. It will perform
!inverse matrix multiplication and really fancy
!error propagation to get the barcode and how close
!the barcode is to the intended target

```

```

subroutine ampoulecalc(filenumber,batchconcentrations,&
batchconcentrationserror,nbatches,ampoules,&
ampouleserror,batchnames,atomcont,requestedat)
use mody
implicit none
!imported variables
integer,intent(in)::filenumber
integer,intent(in)::nbatches
!array for storing the concentrations of batches in aliquots
real(8),allocatable,intent(out)::ampoules(:)
real(8),allocatable,intent(out)::ampouleserror(:)
real(8),intent(in)::batchconcentrations(:, :)
real(8),intent(in)::batchconcentrationserror(:, :)
real(8),intent(in)::atomcont(:, :) !atomic concentrations of all
isotopes in input file
integer,intent(in)::requestedat(:) !location of batch isotopes
in input file
character(*),intent(in)::batchnames(:, :)

!subroutine specific variables

```

```

real(8)::A(nbatches,nbatches) !for the a matrix in least squares
  subroutine
real(8)::dA(nbatches,nbatches) !error in A
real(8)::normalizingfactors(nbatches)
!the mass and atomic concentrations of each isotope from the
  gamma spectroscopy
real(8)::b(nbatches)
real(8)::db(nbatches) !error in b
!bounding matrix to keep answers realistic. 0 is lower bound
  and 1 is upper bound.
real(8)::bl(nbatches),bu(nbatches)
real(8)::x(nbatches)
real(8)::oldx
real(8)::dx(nbatches) !error in x
real(8)::relerror,ampoulesum
real(8),parameter::epis=1d-6
character(1)::choice
integer::i

print *,"Preparing data to unfold the barcode"
print *

!allocate array
allocate(ampoules(nbatches),ampouleserror(nbatches))

!transpose batchconcentrations matrix to be

```

```

!ready for subroutine
A=transpose(batchconcentrations)
dA=transpose(batchconcentrationserror)

!normalize to make up for missing isotopes
choice="n"
select case(choice)
case("1","one","Y","y","YES","yes")
normalizingfactors=sum(A,1)
do i=1,nbatches
A(:,i)=A(:,i)/normalizingfactors(i)
end do
end select

!bounding matrix to prevent negative answers or answers greater
than 100%
bl(1:nbatches)=0d0 !set lower bound at 0%

bu(1:nbatches)=1d2 !set upper bound to 100%

!fill matrix b with atomic concentration and error
do i=1,nbatches
if(requestedat(i)/=0) then
b(i)=atomcont(1,requestdat(i))
db(i)=atomcont(2,requestdat(i))

```

```

else
b(i)=0d0
db(i)=0d0
end if
end do

print *,"Solving system of equations using matrix inverse method"
print *

!systemequationsolver(A(nbatches,nbatches),B(nbatches),
answers(nbatches),&
!lowerbound(nbatches),upperbound(nbatches))
call systemequationsolver(A,b,x,bl,bu)

!normalize answers
ampoulesum=sum(x)
do i=1,nbatches
oldx=x(i)
x(i)=(x(i)/ampoulesum)*1d2
dx(i)=dx(i)*x(i)/oldx
end do

!calculate error of ampoules
call systemequationerror(A,dA,b,db,x,dx,relerror)

!copy answers to ampoules array 1:nbatches

```

```

ampoules(:)=x(:)
ampouleserror(:)=dx(:)

!adjust ampoules to take into account
!any rounding near the edges
print *,"would you like to round answers outside"
print *,"of the 0-100 range to make logical sense?"
print *
print *,"1 - yes"
print *,"2 - no"
read *,choice
select case(choice)
case("1","one","Y","y","YES","yes")
do i=1,nbatches
if(ampoules(i)<epis) then
print *,"rounding done for ampoule"
print *,i
print *,"old value=",ampoules(i)
ampoules(i)=0d0
print *,"new value=",ampoules(i)
end if
if(ampoules(i)>1d2+epis) then
print *,"rounding done for ampoule"
print *,i
print *,"old value=",ampoules(i)
ampoules(i)=1d2

```

```

print *,"new value=",ampoules(i)
end if
end do
end select

print *,"Ampoules contain the following concentrations of batches
  in atom %"
print *

write(filenummer,*)

print 101
write(filenummer,201) !header

do i=1,nbatches
print 102,adjustl(batchnames(1,i)),&
ampoules(i),ampouleserror(i)
write(filenummer,202) &
adjustl(batchnames(1,i)),&
ampoules(i),ampouleserror(i)
end do
print *
write(filenummer,*)

print *,"Relative Error in system (%)=",relerror
print *

```

```

write(filenum,203) relerror
write(filenum,*)

101 format("Batch",t20,"Concentration (%)",t40,"+- Error (%)")
!header
102 format(a20,f6.2,"%",t45,f6.2,"%") !ampoule information
201 format('; "Batch"; "Concentration (%)"; "Error (%)")' !header
202 format('; ',a30,' "; ',f6.2," "; ',f6.2) !ampoule information
203 format('; ', "Relative Error of system(%)", ' "; ',f14.2)

return
end subroutine

!this will generate a list of possible barcodes
!that could match the calculated ampoule ratios

subroutine ampoulematch(inputfilenum, outputfilenum, &
    ampoules, ampouleserror, nbatches, batchnames)
use mody
implicit none
integer, intent(in)::inputfilenum, outputfilenum
real(8), intent(in)::ampoules(:)
real(8), intent(in)::ampouleserror(:)
integer, intent(in)::nbatches
character(*), intent(in)::batchnames(:, :)

```

```

!subroutine specific
integer::i,j
real(8),allocatable::interval(:),tempcode(:),radiusvector(:)
integer,allocatable::icmin(:)
real(8),allocatable::normarray(:),tempnormarray(:)
integer,allocatable::barcodeintegers(:,:),
tempbarcodeintegers(:,:)
integer::nbarcodes,maxcombos,npossiblebarcodes
real(8)::tempreal1
real(8)::specrad,maxradius
character(80),allocatable::outputarray(:,:)
integer::tempint1,tempint2
logical::inrange,isdone
real(8)::maxinterval=1d2
real(8)::mininterval=1d-2
real(8)::sigma,lowrange,highrange
real(8)::epis=1d-6

print *,"Input file where intervals and sigmas can be found?"
call filenameassign(inputfilenumber)
print *

!read input file and extract intervals and sigmas
!subroutine csvfilereader(outputarray,filenumber,&
!nlines,maxcol,delimiter,textquotes)
call csvfilereader(outputarray,inputfilenumber,tempint1,tempint2,

```



```

";",''')
close(inputfilenumber)

!check number of batches
if(nbatches==1) then
print *, "I'm going to stop right now and declare that since
  there is"
print *, "only 1 degree of freedom, there isn't a way to
  generate"
print *, "different combinations of only 1 thing"
stop
end if

allocate(interval(nbatches),tempcode(nbatches))
allocate(radiusvector(nbatches))

!assign batch enrichment intervals and sigmas
do i=1,nbatches
read(outputarray(i+1,2),*) tempreal1 !intervals
interval(i)=tempreal1
end do

!determine maximum number of barcodes
maxcombos=1
do i=1,nbatches
maxcombos=maxcombos*(nint(1d2/interval(i))+2)

```

```

end do

!allocate arrays
allocate(barcodeintegers(maxcombos,nbatches))
allocate(tempbarcodeintegers(maxcombos,nbatches))
allocate(normarray(maxcombos),tempnormarray(maxcombos))
allocate(icmin(nbatches))

!initialize norms so finding the smallest is easier
tempnormarray(:)=huge(0d0)

!assign sigma value
read(outputarray(2,4),*) tempreal1
sigma=tempreal1
if((sigma<mininterval).or.(sigma>maxinterval)) then
print *,"Sigma value"
print *,"is out of bounds"
print *,"minimum interval=",mininterval
print *,"maximum interval=",maxinterval
stop
end if

!check to make sure that batchnames match up
!and put them in the correct order
do i=1,nbatches
if((trim(adjustl(batchnames(1,i))))/=trim(adjustl

```

```

(outputarray(i+1,1)))) then
print *, "Batch names and order of batch names"
print *, "in interval file and batch"
print *, "information file do not match up"
stop
end if
end do

!check to make sure intervals are within bounds
do i=1,nbatches
if((interval(i)<mininterval).or.(interval(i)>maxinterval))
then
print *, "Interval"
print *, i
print *, "is out of bounds"
print *, "minimum interval=", mininterval
print *, "maximum interval=", maxinterval
stop
end if
end do

!write batch heading information
write(outputfilenumber,*) ';"Barcode Section"'
write(outputfilenumber, '(";")', advance="no")
do i=1,nbatches
write(outputfilenumber,101,advance="no") batchnames(1,i)

```

```

end do

write(outputfilenumber,*) '"Whole Barcode";"Norm"'

!print intervals
write(outputfilenumber,'(";")',advance="no")

do i=1,nbatches
tempint1=int(interval(i)*1d2)
write(outputfilenumber,102,advance="no") tempint1
end do

write(outputfilenumber,*) !go to next line


!create temporary first barcode
do i=1,nbatches
tempcode(i)=0d0
end do


!determine maximum spectral radius
maxradius=0d0
do i=1,nbatches
maxradius=maxradius+1d2**2d0
end do

maxradius=dsqrt(maxradius)


print *,"Searching for possible barcodes"
print *

```

```

!determine possible barcodes
nbarcodes=0
npossiblebarcodes=0
isdone=.false.
do
  tempreal1=0d0
  do i=1,nbatches
    tempreal1=tempreal1+tempcode(i)
  end do
  if(tempreal1>(1d2+epis)) then
    !current digit is too high, reset digit to 0
    !and increase previous digit
    tempcode(nbatches)=0d0
    tempcode(nbatches-1)=tempcode(nbatches-1)+interval
    (nbatches-1)
    do j=nbatches,2,-1
      if(tempcode(j)>1d2) then
        tempcode(j)=0d0
        tempcode(j-1)=tempcode(j-1)+interval(j-1)
      end if
    end do
    if(tempcode(1)>1d2) then
      !all barcodes have been evaluated
      isdone=.true.
    end if
  end do
  else if(tempreal1<(1d2-epis)) then

```

```

!barcode is too low, interval last digit
do j=nbatches,1,-1
if(tempcode(j)<(1d2+epis)) then
tempcode(j)=tempcode(j)+interval(j)
exit
end if
end do
else
!possibly hit a barcode, evaluate further
npossiblebarcodes=npossiblebarcodes+1
inrange=.true.
do j=1,nbatches
lowrange=ampoules(j)-sigma*ampouleserror(j)
highrange=ampoules(j)+sigma*ampouleserror(j)
if((tempcode(j)<lowrange).or.
(tempcode(j)>highrange)) then
inrange=.false.
exit
end if
end do
if(inrange) then !record barcode if it's good
nbarcodes=nbarcodes+1
do j=1,nbatches
tempint1=nint(tempcode(j)*1d2)
tempbarcodeintegers(nbarcodes,j)=tempint1
end do

```

```

!determine spectral distance
radiusvector(:)=tempcode(:)-ampoules(:)
call norm(radiusvector,2,specrad)
tempnormarray(nbarcodes)=(specrad/maxradius)*1d2
end if

!go up to next barcode
do j=nbatches,2,-1
  if(tempcode(j)<(1d2+epis)) then
    tempcode(j)=tempcode(j)+interval(j)
  exit
end if
end do
end if

if(isdone) exit
end do

!reorder barcodes
do i=1,nbarcodes
  icmin=minloc(tempnormarray)
  !place in top spot and erase from list
  normarray(i)=tempnormarray(icmin(1))
  barcodeintegers(i,:)=tempbarcodeintegers(icmin(1),:)
  !erase number
  tempnormarray(icmin(1))=huge(1d0)
  tempbarcodeintegers(icmin(1),:)=huge(1)
end do

```

```

!output barcodes into file
do i=1,nbarcodes
write(outputfilenumber,'(";")',advance="no")
do j=1,nbatches
write(outputfilenumber,102,advance="no")
barcodeintegers(i,j)
end do

!all together
write(outputfilenumber,103,advance="no")
do j=1,nbatches
write(outputfilenumber,104,advance="no")
barcodeintegers(i,j)
end do
write(outputfilenumber,105,advance="no") !finish up whole
barcode

write(outputfilenumber,106) normarray(i)
!record distance between point and barcode
end do

!output number of barcodes
print *
print *,"There are"
print *,nbarcodes

```



```

print *,"possible barcodes"
print *,"out of a total of"
print *,npossiblebarcodes
print *

write(outputfilenumber,*)
write(outputfilenumber,107) nbarcodes
write(outputfilenumber,108) npossiblebarcodes
write(outputfilenumber,*)

if(allocated(interval)) deallocate(interval)
if(allocated(tempcode)) deallocate(tempcode)
if(allocated(radiusvector)) deallocate(radiusvector)
if(allocated(barcodeintegers)) deallocate(barcodeintegers)
if(allocated(radiusvector)) deallocate(radiusvector)
if(allocated(tempbarcodeintegers)) deallocate
(tempbarcodeintegers)
if(allocated(normarray)) deallocate(normarray)
if(allocated(tempnormarray)) deallocate(tempnormarray)
if(allocated(icmin)) deallocate(icmin)
deallocate(outputarray)

print *,"finished finding possible barcodes"
print *

101 format('"',a15,'"';')

```

```

102 format(' ',i5.5,'";')
103 format(' ')
104 format(i5.5)
105 format('";')
106 format(f6.2)
107 format('; "Number of barcodes"; ',i15)
108 format('; "Number of total barcodes"; ',i15)

return

end subroutine

!this will calculate the distance between two
!isotopic vectors

function barcodedistance(set1,set2)
use mody
implicit none
real(8)::barcodedistance
real(8),intent(in)::set1(:),set2(:)

!subroutine specific
integer::set1size,set2size,nbatches
integer::i
real(8),allocatable::radiusvector(:)
real(8)::specrad,maxradius

!determine number of batches and make sure they match

```

```

set1size=size(set1,1)
set2size=size(set2,1)

if(set1size/=set2size) then
print *, "error in barcodedistance"
print *, "the size of the barcodes do not match"
stop
end if

nbatches=set1size

allocate(radiusvector(nbatches))

!determine maximum spectral radius
maxradius=0d0
do i=1,nbatches
maxradius=maxradius+1d2**2d0
end do
maxradius=dsqrt(maxradius)

!determine spectral distance
radiusvector(:)=set1(:)-set2(:)
call norm(radiusvector,2,specrad)
barcodedistance=(specrad/maxradius)*1d2

deallocate(radiusvector)

```

```

end function

!this will generate a list of possible barcodes given certain
  isotopes and enrichment intervals

subroutine barcodegenerator(inputfilenumber,outputfilenumber)
use mody
implicit none
integer,intent(in)::inputfilenumber,outputfilenumber

!subroutine specific
integer::nbarcodes,nbatches
integer::i,j
logical::isdone=.false.
real(8),allocatable::interval(:),tempcode(:)
real(8)::tempreal1
integer::tempint1,tempint2
character(80),allocatable::batchname(:)
character(80),allocatable::outputarray(:, :)
real(8)::maxinterval=1d2
real(8)::mininterval=1d-2
real(8)::epis=1d-6

print *, "Name of input file?"
call filenameassign(inputfilenumber)
print *

```

```

!subroutine csvfilereader(outputarray,filenumber,&
!nlines,maxcol,delimiter,textquotes)
call csvfilereader(outputarray,inputfilenumber,tempint1,
tempint2,";",'")')
close(inputfilenumber)

!count number of batches
nbatches=tempint1-1

!check number of batches
if(nbatches==1) then
print *, "I'm going to stop right now and declare that since
there is"
print *, "only 1 degree of freedom, there isn't a way to
generate"
print *, "different combinations of only 1 thing"
stop
end if

!allocate arrays
allocate(interval(nbatches),tempcode(nbatches))
allocate(batchname(nbatches))

!assign batch enrichment names
do i=1,nbatches

```

```

batchname(i)=trim(adjustl(outputarray(i+1,1)))
end do

!assign batch enrichment intervals and sigmas
do i=1,nbatches
read(outputarray(i+1,2),*) tempreal1
interval(i)=tempreal1
end do

!check to make sure intervals are within bounds
do i=1,nbatches
if((interval(i)<mininterval).or.(interval(i)>maxinterval))
then
print *,"Interval"
print *,i
print *,"is out of bounds"
print *,"minimum interval=",mininterval
print *,"maximum interval=",maxinterval
stop
end if
end do

!write batch heading information
write(outputfilenumber,'(";")',advance="no")
do i=1,nbatches
write(outputfilenumber,101,advance="no") batchname(i)

```

```

end do

write(outputfilenumber,*) ';"Whole Barcode";'

!print intervals
write(outputfilenumber,'(";")',advance="no")
do i=1,nbatches
tempint1=int(interval(i)*1d2)
write(outputfilenumber,102,advance="no") tempint1
print *,batchname(i),interval(i)
end do

write(outputfilenumber,*) !go to next line
write(outputfilenumber,*) !go to next line

!create temporary first barcode
do i=1,nbatches
tempcode(i)=0d0
end do

!determine possible barcodes
nbarcodes=0
do
tempreal1=0d0
do i=1,nbatches
tempreal1=tempreal1+tempcode(i)
end do

```

```

if(tempreal1>(1d2+epis)) then
!current digit is too high, reset digit to 0
!and increase previous digit
tempcode(nbatches)=0d0
tempcode(nbatches-1)=tempcode(nbatches-1)+interval
(nbatches-1)

do j=nbatches,2,-1
if(tempcode(j)>1d2) then
tempcode(j)=0d0
tempcode(j-1)=tempcode(j-1)+interval(j-1)
end if
if(tempcode(1)>1d2) then
!all barcodes have been evaluated
isdone=.true.
end if
end do

else if(tempreal1<(1d2-epis)) then
!barcode is too low, interval last digit
do j=nbatches,1,-1
if(tempcode(j)<(1d2+epis)) then
tempcode(j)=tempcode(j)+interval(j)
exit
end if
end do
else

```



```

!hit a magic barcode, record it
nbarcodes=nbarcodes+1
write(outputfilenumber,'(";")',advance="no")
do j=1,nbatches
tempint1=nint(tempcode(j)*1d2)
write(outputfilenumber,102,advance="no") tempint1
end do

!all together
write(outputfilenumber,103,advance="no")
do j=1,nbatches
tempint1=nint(tempcode(j)*1d2)
write(outputfilenumber,104,advance="no") tempint1
end do

write(outputfilenumber,105) !finish and go to next line
!go up to next barcode
do j=nbatches,2,-1
if(tempcode(j)<(1d2+epis)) then
tempcode(j)=tempcode(j)+interval(j)
exit
end if
end do
end if
if(isdone) exit
end do

!output number of barcodes

```

```

print *
print *, "There are"
print *, nbarcodes
print *, "barcodes"
print *

write(outputfilenumber,*)
write(outputfilenumber,106) nbarcodes

close(outputfilenumber)

deallocate(interval,tempcode,outputarray)

print *, "barcode generator complete"

101 format(' ',a15,'";')
102 format(' ',i5.5,'";')
103 format('; "')
104 format(i5.5)
105 format('";')
106 format('; "Number of barcodes";',i15)

return

end subroutine

!this will calculate the amount of material required from each batch

```

```

to
!form a specific atom ratio of batches and make a barcode
!
!many subroutines used are not used for their original purpose.
Some variables
!are dummies just for the purpose of the subroutines even if they
aren't used.

subroutine barcodemass(inputfilenumber,outputfilenumber,
batchfilenumber)
use mody
implicit none
integer,intent(in)::inputfilenumber !file to read oxides from
integer,intent(in)::outputfilenumber !output file number where
prescribed masses are output to
integer,intent(in)::batchfilenumber !file where batch
information is stored

!specific variables
integer :: i,j
real(8),allocatable::amatrix(:,:),xmatrix(:),bmatrix(:)
real(8),allocatable::nuclideinfo(:),masspercentage(:)
real(8),allocatable::measuredmass(:),measuredmassfraction(:)
real(8),allocatable:: massratio(:)
integer::nisotopes !number of isotopes in input file
integer::nbatches !number of batches read from ratio text file

```

```

character(80)::sampledescription
real(8)::samplesize
character(80)::tempchar1,tempchar2,tempchar3
real(8)::tempreal1
integer::tempint1,tempint2
integer::noxides
logical::foundinoxidelist
character(80)::barcodestring
real(8),allocatable::batchconcentrations(:,,:)
!stores the concentration of batches of isotopes
character(80),allocatable::batchnames(:,,:)
!stores the names or id tags of the stock batch isotopes
real(8),allocatable::batchconcentrationerror(:,,:)
character(80),allocatable::inputarray(:,,:)
integer,parameter::offset=1

!read desired barcode ratios from terminal

!read sample description
print *,"Sample description"
print *,"If more than one word, put in quotes"
read *, sampledescription
print *

!put description in outputfile
print *,"Sample description=",sampledescription

```

```

print *
write(outputfilenumber,*) ';;',trim(adjustl(sampldescription))
,',';

!Enter sample mass
print *, "Enter sample mass with a space and then mass unit"
read *,tempchar1,tempchar2
read(tempchar1,*) tempreal1
samplesize=massconvert(tempreal1,tempchar2,"g") !convert to grams
write(outputfilenumber,101) tempreal1,tempchar2

print *, "Sample mass in grams"
print '(en14.6)',samplesize
print *

!read barcode
print *, "Please enter barcode all in one number without spaces"
read *,tempchar1
print *
barcodestring=trim(adjustl(tempchar1))
tempint1=len_trim(tempchar1)/5 !each barcode is 5 digits long

!check to make sure barcode is multiples of 5
if(tempint1*5/=len_trim(tempchar1)) then
print *, "Barcode length should be a multiple of 5 units"
stop

```

```

end if
write(outputfilenumber,*)
write(outputfilenumber,*) ';"Barcode"'
write(outputfilenumber,103) trim(adjustl(tempchar1))
write(outputfilenumber,*)

nisotopes=tempint1

!allocate arrays
if(.not.allocated(nuclideinfo)) allocate(nuclideinfo(nisotopes))

!set default variables
do i=1,nisotopes
nuclideinfo(i)=0d0
end do

!read barcode into arrays
!parse barcode into atom% of each nuclide
do i=1,nisotopes
tempint1=((i-1)*5)+1
tempint2=tempint1+4
tempchar1=barcodestring(tempint1:tempint2)
read(tempchar1,*) tempreal1
nuclideinfo(i)=tempreal1/1d2 !barcode number in atom%
end do

```

```

!make sure the barcode adds up to 100%
tempreal1=sum(nuclideinfo)-1d2
if((tempreal1>1d-4).or.(tempreal1<-1d-4)) then
print *, "Barcode is invalid because it doesn't"
print *, "add up to 100%"
stop
end if

print *, "Nuclides=", nisotopes
print *

print *, "Done reading input file"
print *

!allocate matrix
if(.not.allocated(bmatrix)) allocate(bmatrix(nisotopes))
if(.not.allocated(massratio)) allocate(massratio(nisotopes))
if(.not.allocated(masspercentage)) allocate(masspercentage
(nisotopes))
if(.not.allocated(measuredmass)) allocate(measuredmass
(nisotopes))
if(.not.allocated(measuredmassfraction)) then
allocate(measuredmassfraction(nisotopes))
end if

!initialize arrays

```

```

measuredmassfraction(:)=1d0

!read gravimetric list to find out weighted atomic masses for
  each batch
print *,"Name of file containing a list of batches"
print *,"and their associated atomic weights and"
print *,"gravimetric information?"
print *,"column 1=batch name like CD-106 102123"
print *,"column 2=batch atomic weight"
print *,"column 3=gravimetric fraction by weight"
call filenameassign(inputfilenumber)
call csvfilereader(inputarray,inputfilenumber,tempint1,
tempint2,";",',')
close(inputfilenumber)

noxides=tempint1-offset

!read batch information
call batchread(batchfilenumber,2,batchconcentrations,&
batchconcentrationserror,batchnames,nbatches)

!search for matches
do i=1,nisotopes
foundinoxidelist=.false.
do j=1,noxides
tempchar1=trim(adjustl(inputarray(j+offset,1)))

```



```

tempchar2=trim(adjustl(inputarray(j+offset,2)))
tempchar3=trim(adjustl(inputarray(j+offset,3)))
if(tempchar1==batchnames(1,i)) then
foundinoxidelist=.true.
if(isnumeric(tempchar2)) then
read(tempchar2,*) tempreal1
bmatrix(i)=tempreal1
else
print *,"error in barcodemass"
print *,"expecting numerical value for atomic
weight"
print *,"in reading file for batch"
print *,tempchar1
stop
end if
if(isnumeric(tempchar3)) then
read(tempchar3,*) tempreal1
measuredmassfraction(i)=tempreal1/1d2
else
print *,"error in barcodemass"
print *,"expecting numerical value for
gravimetric value"
print *,"in reading file for batch"
print *,tempchar1
stop
end if

```

```

exit
end if
end do

if(.not.foundinoxidelist) then
print *, "error in barcode mass"
print *, "a weighted atomic mass and gravimetric"
print *, "information were not found for batch"
print *, batchnames(1,i)
stop
end if
end do

!translate atom% to mass%
call atommassconvert(1,nuclideinfo,bmatrix,masspercentage)
massratio(:)=masspercentage(:)/1d2

!adjust for dilute materials
measuredmass(:)=massratio(:)*samplesize/measuredmassfraction(:)

!output mass readings to output file
write(outputfilenumber,*) ';;"mass (g)";"mass fraction (%)"'
do i=1,nbatches
write(outputfilenumber,102) batchnames(1,i),measuredmass(i),
masspercentage(i)
end do
write(outputfilenumber,*)

```

```

write(outputfilenumber,*)
write(outputfilenumber,*)

!output multiplicative matrix
!write header
write(outputfilenumber,201) "To go from column to row,
  multiply by"
write(outputfilenumber,'(";;")',advance="no")
do i=1,nbatches
write(outputfilenumber,202,advance="no") &
trim(adjustl(batchnames(1,i)))
end do
write(outputfilenumber,*)

do i=1,nbatches
write(outputfilenumber,203,advance="no") &
trim(adjustl(batchnames(1,i)))
do j=1,nbatches
if((massratio(i)/=0d0).and.(massratio(j)/=0d0)) then
tempreal1=massratio(j)/massratio(i)
write(outputfilenumber,204,advance="no") tempreal1
else
write(outputfilenumber,204,advance="no") 0d0
end if
end do
write(outputfilenumber,*)

```

```

end do

!close output file
close(outputfilenumber)

!Deallocate all arrays
if(allocated(batchconcentrations))
    deallocate(batchconcentrations)
if(allocated(batchconcentrationserror)) then
    deallocate(batchconcentrationserror)
end if
if(allocated(batchnames)) deallocate(batchnames)
if(allocated(nuclideinfo)) deallocate(nuclideinfo)
if(allocated(amatrix)) deallocate(amatrix)
if(allocated(xmatrix)) deallocate(xmatrix)
if(allocated(bmatrix)) deallocate(bmatrix)
if(allocated(massratio)) deallocate(massratio)
if(allocated(masspercentage)) deallocate(masspercentage)
if(allocated(measuredmass)) deallocate(measuredmass)
if(allocated(measuredmassfraction)) deallocate
(measuredmassfraction)
deallocate(inputarray)

print *, "Program Complete"

101 format(';"Total sample mass";',f12.8,',','"',a3,',";')

```

```

102 format(';',a20,'"',f10.5,"",f6.2)
103 format(';',a,'"')
201 format(';',a40,'"')
202 format('"',a,'";')
203 format(';',a,'";')
204 format(f12.8,"")

return

end subroutine barcodemass

subroutine batchread(filenummer,batchnamescategories,&
batchconcentrations,batchconcentrationserror,&
batchnames,nbatches)

use mody

implicit none

integer,intent(in)::filenummer !unit number of text file
containing batch information

integer,intent(in)::batchnamescategories !number of categories
in the batchnames array

integer,intent(inout)::nbatches !number of batches read from
ratio text file

!stores the concentration of batches of isotopes
real(8),allocatable,intent(inout)::batchconcentrations(:, :)

!stores error of batch concentrations
real(8),allocatable,intent(inout)::batchconcentrationserror(:, :)

!stores the names or id tags of the stock batch isotopes
character(*),allocatable,intent(inout)::batchnames(:, :)

```

```

!subroutine specific
integer::nbatchisotopes
character(80),allocatable::temparray(:, :)

!The usual suspects
integer ::tempint1,tempint2
integer ::i,j
real(8) ::tempreal1

!open csv file containing batch information to read from
print *, "Name of file containing batch information?"
call filenameassign(filename)
print *

!read csv file into a character array
call csvfilereader(temparray,filename,tempint1,tempint2,
";",',','"')

close(filename) !Close ratio file

!get number of batches
nbatches=tempint1-1
nbatchisotopes=tempint2-1

print *, "Number of batches found="

```

```

print *,nbatches
print *
print *,"Number of isotopes in each batch="
print *,nbatchisotopes
print *

!verify that isotopes matches the number of batches to form a
  well posed problem
if(nbatchisotopes>nbatches) then
print *,"Too many isotopes"
print *,"This forms an overdetermined system"
stop
else if(nbatchisotopes<nbatches) then
print *,"Not enough isotopes"
print *,"This forms an underdetermined system"
stop
end if

!allocate arrays dealing with batches
allocate(batchconcentrations(nbatches,nbatchisotopes))
allocate(batchnames(batchnamescategories,nbatches))
allocate(batchconcentrationerror(nbatches,nbatchisotopes))

!transfer names of batches
batchnames(1,1:nbatches)=temparray(2:nbatches+1,1)

```

```

!transfer names of isotopes in batches
batchnames(2,1:nbatchisotopes)=temparray(1,2:nbatchisotopes+1)

!transfer batch atom% assay
do i=1,nbatches
do j=1,nbatchisotopes
read(temparray(i+1,j+1),*) tempreal1
batchconcentrations(i,j)=tempreal1
end do
end do

!read error in batch concentrations file
print *,"Name of file containing"
print *,"error in batch information?"
call filenameassign(filename)
print *

!read csv file into a character array
deallocate(temparray)
call csvfilereader(temparray,filename,tempint1,tempint2,
";",',','"')

close(filename) !Close error file

!transfer error in batch atom% assay
do i=1,nbatches

```



```

do j=1,nbatchisotopes
read(temparray(i+1,j+1),*) tempreal1
batchconcentrationerror(i,j)=tempreal1
end do
end do

deallocate(temparray)

return
end subroutine batchread

!This will calculate the atomic and mass fractions given the
activity
!and activity uncertainty from NAA. It will also calculate all of
!the associated uncertainty

subroutine concentrationcalculate(avogadro,avogadrouncert,atomcont,
nisotopes,&
librarynumbers,elementconc,samplesize,&
samplesizeuncert,irradiationtime,
irradiationtimeuncertainty,&
thermalflux,thermalfluxuncert,fastflux,fastfluxuncert,&
isomass,isomassuncert,masscont,natoms,natomsuncert,&
librarylocation,activityinfo,activityuncertainty,&
activatedmass,activatedmassuncert,&
standardnumbers,standardavailable)

```

```

implicit none

!imported variables

real(8),intent(in)::avogadro !avogadro constant
real(8),intent(in)::avogadrouncert !error in avogadro constant
integer,intent(in)::nisotopes !number of isotopes in sample
real(8),intent(in)::librarynumbers(:, :) !Data read from the
    library file
real(8),intent(in)::samplesize,samplesizeuncert !whole sample
    mass
real(8),intent(in)::irradiationtime !time of irradiation
real(8),intent(in)::irradiationtimeuncertainty !error in time
    of irradiation
real(8),intent(in)::thermalflux,fastflux !reactor fluxes
real(8),intent(in)::thermalfluxuncert,fastfluxuncert !flux
    uncertainties
integer,intent(in)::librarylocation(:) !location of input
    entries in library
logical,intent(in)::standardavailable(:) !If to use comparator
    method or not
real(8),allocatable,intent(in)::standardnumbers(:, :)
real(8),allocatable,intent(in)::activityinfo(:) !average activity
    of each isotope
real(8),allocatable,intent(in)::activityuncertainty(:)
!activity uncertainty of each isotope
real(8),allocatable,intent(inout)::natoms(:)
!number of atoms of each isotope in input file

```

```

real(8),allocatable,intent(inout)::natomsuncert(:) !uncertainty
  of natoms(:)
real(8),allocatable,intent(inout)::isomass(:) !mass of each
  isotope in input file
real(8),allocatable,intent(inout)::isomassuncert(:) !uncertainty
  of isotopic mass
real(8),allocatable,intent(inout)::masscont(:, :)
!mass fraction of each isotope in input file
real(8),allocatable,intent(inout)::atomcont(:, :)
!atom fraction of each isotope in input file
real(8),allocatable,intent(inout)::elementconc(:, :)
!elemental concentrations and uncertainty
real(8),intent(out)::activatedmass,activatedmassuncert
!mass of activated material and uncertainty

!The usual suspects
integer::i,j
real(8)::tempreal1,tempreal2,tempreal3,tempreal4
real(8)::tempreal5,tempreal6,tempreal7

!subroutine specific variables
real(8)::totalatoms,totalatomsuncert !number of atoms in entire
  sample and uncertainty
real(8)::thermalxs,thermalxsuncert,fastxs,fastxsuncert !cross
  sections
real(8)::activity,actuncert

```

```

real(8)::atomicmass,atomicmassuncert !atomic masses of isotope
real(8)::atomicweight,atomicweightuncert !atomic weight of
    element
real(8)::abundance,abundanceuncert !natural abundance of isotope
real(8)::lambda,lambdauncert !decay constant

print *, "Calculating concentrations"

!allocate arrays to hold concentration information
allocate(isomass(nisotopes),isomassuncert(nisotopes))
allocate(natoms(nisotopes),natomsuncert(nisotopes))
allocate(elementconc(2,nisotopes))
allocate(masscont(4,nisotopes),atomcont(4,nisotopes))

!all mass values are assumed to arrive to this
!point in grams and should be properly converted beforehand

!Determine mass of trace elements
do i=1,nisotopes
    j=librarylocation(i)
    activity=activityinfo(i)
    if(activity==0d0) activity=1 !prevents /0 errors
        calculating uncertainty
    actuncert=activityuncertainty(i)
    thermalxs=librarynumbers(1,j)
    thermalxsuncert=librarynumbers(2,j)

```

```

fastxs=librarynumbers(3,j)
fastxsuncert=librarynumbers(4,j)
lambda=librarynumbers(5,j)
lambdauncert=librarynumbers(6,j)
atomicmass=librarynumbers(7,j)
atomicmassuncert=librarynumbers(8,j)
atomicweight=librarynumbers(9,j)
atomicweightuncert=librarynumbers(10,j)
abundance=librarynumbers(11,j)
abundanceuncert=librarynumbers(12,j)

```

!There are 2 methods of calculating things. The first is
using

!the absolute method involving flux values and cross
sections

!The other involves using a comparator sample of known
properties

!to compare the sample against

!Parametric method

!number of atoms=(Activity/((thermalxsection*Thermflux+
resonanceintegral*fastflux)

!*(1.0-EXP(0-lambda*time))))

!natoms is the number of target atoms in the sample

!isomass=(Natoms*atomic mass)/avogrado number

!masscont(1,:)=(mass of isotope/mass of samplesize)

```

!masscont(2,:)=uncertainty of masscont(1,:)
!masscont(3,:)=(mass of isotope)/(total mass of
  radioisotopes)
!masscont(4,:)=uncertainty of masscont(3,:)
!atomcont(1,:)=natoms/sum of all natoms
!atomcont(2,:)=uncertainty of atomcont(1,:)

!Comparative method
!natoms=((Activity sample)/(Activity standard))*&
! (mass of isotope in standard)*(avogadro/atomic mass)

if(standardavailable(i)) then !use comparative NAA
natoms(i)=(activity/standardnumbers(i,13))*&
  (standardnumbers(i,11)*avogadro)/atomicmass
natomsuncert(i)=natoms(i)*dsqrt(&
  (actuncert/activity)**2d0+&
  (standardnumbers(i,14)/standardnumbers(i,13))**2d0+&
  (standardnumbers(i,12)/standardnumbers(i,11))**2d0+&
  (avogadrouncert/avogadro)**2d0+&
  (atomicmassuncert/atomicmass)**2d0)

!isomass=activity/(standard activity)*&
!(mass of isotope in standard)
isomass(i)=(activity/standardnumbers(i,13))*&
  standardnumbers(i,11)
isomassuncert(i)=isomass(i)*dsqrt(&

```

```

(actuncert/activity)**2d0+&
(standardnumbers(i,14)/standardnumbers(i,13))**2d0+&
(standardnumbers(i,12)/standardnumbers(i,11))**2d0)
else !use parametric NAA
natoms(i)=activity/((thermalxs*thermalflux+fastxs*
fastflux)&
*(1d0-dexp((-lambda)*irradiationtime)))

!find uncertainty in number of atoms
!This comes from taking the partial differentials on
!natoms=activity/((sigmathermal*phithermal+sigmafast*
phifast)*(1-exp(-lambda*time)))
!using the traditional differential error propagation
formula
!there are 7 terms for each of the 7 variables
tempreal1=actuncert**2d0/((thermalxs*thermalflux+&
fastxs*fastflux)**2d0*(1d0-dexp((-lambda)*
irradiationtime))**2d0)
tempreal2=(thermalfluxuncert**2d0*thermalxs**2d0*
activity**2d0)/&
((thermalxs*thermalflux+fastxs*fastflux)**4d0*&
(1d0-dexp((-lambda)*irradiationtime))**2d0)
tempreal3=(fastfluxuncert**2d0*fastxs**2d0*activity**
2d0)/&
((thermalxs*thermalflux+fastxs*fastflux)**4d0*&
(1d0-dexp((-lambda)*irradiationtime))**2d0)

```

```

tempreal4=(thermalxsuncert**2d0*thermalflux**2d0*
activity**2d0)/&
((thermalxs*thermalflux+fastxs*fastflux)**4d0*&
(1d0-dexp((-lambda)*irradiationtime))**2d0)
tempreal5=(fastxsuncert**2d0*fastflux**2d0*activity**
2d0)/&
((thermalxs*thermalflux+fastxs*fastflux)**4d0*&
(1d0-dexp((-lambda)*irradiationtime))**2d0)
tempreal6=(irradiationtimeuncertainty**2d0*lambda**2d0*&
activity**2d0*dexp((-2d0)*lambda*irradiationtime))/&
((thermalxs*thermalflux+fastxs*fastflux)**2d0*&
(1d0-dexp((-lambda)*irradiationtime))**4d0)
tempreal7=(irradiationtime**2d0*lambdauncert**2d0*&
activity**2d0*dexp((-2d0)*lambda*irradiationtime))/&
((thermalxs*thermalflux+fastxs*fastflux)**2d0*&
(1d0-dexp((-lambda)*irradiationtime))**4d0)
natomsuncert(i)=dsqrt(tempreal1+tempreal2+tempreal3+&
tempreal4+tempreal5+tempreal6+tempreal7)

```

```

!find mass of isotope for each gamma line
!mass=(number of atoms)*(atomic mass)/(avogadros)
isomass(i)=(natoms(i)*(atomicmass))/avogadro
isomassuncert(i)=isomass(i)*dsqrt(&
(natomsuncert(i)/natoms(i))**2d0+&
(atomicmassuncert/atomicmass)**2d0+&
(avogadrouncert/avogadro)**2d0)

```



```

end if

!find elemental concentration and uncertainty assuming
  natural abundance
elementconc(1,i)=(natoms(i)/avogadro)/abundance*atomicweight
/samplesize
elementconc(2,i)=elementconc(1,i)*dsqrt(&
(natomsuncert(i)/natoms(i))**2d0+&
(avogadrouncert/avogadro)**2d0+&
(abundanceuncert/abundance)**2d0+&
(atomicweightuncert/atomicweight)**2d0+&
(samplesizeuncert/samplesize)**2d0)
end do

!find mass concentration compared to the whole sample
do i=1,nisotopes
masscont(1,i)=isomass(i)/samplesize !mass concentration
!error in mass concentration
masscont(2,i)=masscont(1,i)*&
dsqrt((isomassuncert(i)/isomass(i))**2d0+&
(samplesizeuncert/samplesize)**2d0)
end do

!find mass of sample that was activated
activatedmass=0d0
activatedmassuncert=0d0

```

```

do i=1,nisotopes
activatedmass=activatedmass+isomass(i)
activatedmassuncert=activatedmassuncert+(isomassuncert(i))
**2d0
end do
activatedmassuncert=dsqrt(activatedmassuncert)

!find mass ratio of sample that was activated
do i=1,nisotopes
masscont(3,i)=isomass(i)/activatedmass !mass fraction
masscont(4,i)=masscont(3,i)*dsqrt((isomassuncert(i)/
isomass(i))**2d0+&
(activatedmassuncert/activatedmass)**2d0)
end do

!sum all atoms and find atomic concentration to be stored in
array
totalatoms=0d0
totalatomsuncert=0d0
do i=1,nisotopes
totalatoms=totalatoms+natoms(i)
totalatomsuncert=totalatomsuncert+natomsuncert(i)
end do
totalatomsuncert=dsqrt(totalatomsuncert)

!atomic fraction of known radioactive isotopes

```

```

do i=1,nisotopes
atomcont(1,i)=natoms(i)/totalatoms
atomcont(2,i)=atomcont(1,i)*dsqrt(&
(natomsuncert(i)/natoms(i))*2d0+&
(totalatomsuncert/totalatoms))*2d0)
end do

!normalize to 100%
atomcont(:,:)=atomcont(:,:)*1d2
masscont(3,:)=masscont(3,:)*1d2
masscont(4,:)=masscont(4,:)*1d2

print *,"Done calculating concentrations"

return

end subroutine concentrationcalculate

!This will output the results of concentration calculate
!into a csv format file

subroutine concentrationoutput(filenummer,atomcont,nisotopes,
elementconc,&
samplesize,samplesizeuncert,isomass,isomassuncert,&
masscont,natoms,natomsuncert,librarylocation,&
targetnucli,&
thermalflux,thermalfluxuncert,&

```

```

fastflux,fastfluxuncert,&
irradiationtime,irradiationtimeuncertainty,&
activatedmass,activatedmassuncert)

implicit none

!imported variables

integer,intent(in)::nisotopes !number of isotopes in sample
real(8),intent(in)::samplesize,samplesizeuncert !whole sample
    mass

real(8),intent(in)::irradiationtime !time of irradiation
real(8),intent(in)::irradiationtimeuncertainty !error in time
of irradiation

real(8),intent(in)::thermalflux,fastflux !reactor fluxes
real(8),intent(in)::thermalfluxuncert,fastfluxuncert !flux
uncertainties

integer,intent(in)::librarylocation(:) !location of input
    entries in library

real(8),allocatable,intent(in)::natoms(:)

!number of atoms of each isotope in input file
real(8),allocatable,intent(in)::natomsuncert(:) !uncertainty
    of natoms(:)

real(8),allocatable,intent(in)::isomass(:) !mass of each isotope
    in input file

real(8),allocatable,intent(in)::isomassuncert(:) !uncertainty of
    isotopic mass

real(8),allocatable,intent(in)::masscont(:, :)

!mass fraction of each isotope in input file

```

```

real(8),allocatable,intent(in)::atomcont(:,,:)
!atom fraction of each isotope in input file
real(8),allocatable,intent(in)::elementconc(:,,:)
!elemental concentrations and uncertainty
real(8),intent(in)::activatedmass,activatedmassuncert
!mass of activated material and uncertainty
character(*),intent(in)::targetnucli(:) !names of activated
nuclides
integer,intent(in)::filenumber !output file number

!The usual suspects
integer :: i

print *,"writing concentration calculations"
print *,"to output file"

write(filenumber,*) !skip line

!mass concentration and mass fraction
write(filenumber,101) !mass concentration header
do i=1,nisotopes
write(filenumber,102) targetnucli(librarylocation(i)),&
masscont(1,i),masscont(2,i),&
masscont(3,i),masscont(4,i)
!1=mass concentration 2=uncertainty
!3=mass %, 4=uncertainty

```

```

end do

write(filenum,*) !skip line


!atom % section
write(filenum,103) !atom concentration header
do i=1,nisotopes
write(filenum,104) targetnucli(librarylocation(i)),&
atomcont(1,i),atomcont(2,i)
end do
write(filenum,*) !skip line


!isotopic mass section
write(filenum,105) !isotopic mass concentration header
do i=1,nisotopes
write(filenum,106) targetnucli(librarylocation(i)),
isomass(i),isomassuncert(i)
end do
write(filenum,*) !skip line


!element concentration section
write(filenum,107) !element conc header
do i=1,nisotopes
write(filenum,108) targetnucli(librarylocation(i)),&
elementconc(1,i),elementconc(2,i)
end do
write(filenum,*) !skip line

```

```

!number of atoms section
write(filenum,109) !Number of atoms header
do i=1,nisotopes
write(filenum,110) targetnucli(librarylocation(i)),&
natoms(i),natomsuncert(i)
end do
write(filenum,*) !skip line

!activated mass section
write(filenum,111) activatedmass,activatedmassuncert
write(filenum,*) !skip line

!sample size section
write(filenum,112) samplesize,samplesizeuncert
write(filenum,*) !skip line

!irradiation time
write(filenum,113) irradiationtime,irradiationtimeuncertainty
write(filenum,*) !skip line

!flux sections
write(filenum,114) thermalflux,thermalfluxuncert
write(filenum,115) fastflux,fastfluxuncert
write(filenum,*) !skip line

```

```

101 format(';"Isotope";"Sample mass concentration";"Sample mass
    concentration uncertainty";',&
&' "Radioisotope mass %";"Radioisotope mass % uncertainty"')
102 format(';"',a7,'";',es14.6,"";es14.6,"";f6.2,"";f6.2)
103 format(';"Isotope";"Atom %";"Atom % uncertainty"')
104 format(';"',a7,'";',f6.2,"";f6.2)
105 format(';"Isotope";"Isotopic mass (g)";"Isotopic mass
    uncertainty"')
106 format(';"',a7,'";',es14.6,"";es14.6)
107 format(';"Isotope";"Element Concentration";"Element
Concentration Uncertainty"')
108 format(';"',a7,'";',es14.6,"";es14.6)
109 format(';"Isotope";"Number of Atoms";"Uncertainty"')
110 format(';"',a7,'";',es14.6,"";es14.6)
111 format(';"Activated mass (g)";',en14.6,"";en14.6)
112 format(';"Sample mass (g)";',en14.6,"";en14.6)
113 format(';"Irradiation time (s)";',es14.6,"";es14.6)
114 format(';"Thermal Flux";',es14.6,"";es14.6)
115 format(';"Fast Flux";',es14.6,"";es14.6)

return

end subroutine concentrationoutput

subroutine libraryread(filenummer,nlibrarysize,nisotopes,&
isotopes,librarylocation,librarynumbers,&
productnucli,targetnucli,converselibrarylocation)

```



```

use mody

implicit none

integer,intent(in)::filenumber !unit number of library text file

integer,intent(inout)::nlibrarysize !defines how many nuclides
are in library

character(*),allocatable,intent(inout)::productnucli(:)

!Names of product nuclides from activation according to library

character(*),allocatable,intent(inout)::targetnucli(:)

!Names of target nuclides collected from library

integer,allocatable,intent(inout)::converselibrarylocation(:)

!Location of library isotopes in input file

real(8),allocatable,intent(inout)::librarynumbers(:, :)

integer,intent(in)::nisotopes !number of isotopes in input file

character(*),intent(in)::isotopes(:)

integer,allocatable,intent(inout)::librarylocation(:)

!location of input nuclides in library


!subroutine specific

character(40),allocatable::outputarray(:, :)

integer,parameter::nlibrarynumberscategories=12

integer,parameter::offset=1


!The usual suspects

integer::i,j

integer::tempint1,tempint2

real(8)::tempreal1,tempreal2,tempreal3,tempreal4,tempreal5

```

```

real(8)::tempreal6,tempreal7,tempreal8,tempreal9,tempreal10
real(8)::tempreal11,tempreal12

!read library information
print *,"Reading library file"
print *
rewind(filenummer)
call csvfilereader(outputarray,filenummer,&
tempint1,tempint2,";",'")
close(filenummer)

!size everything
nlibrarysize=tempint1-offset
if(tempint2>20) then
print *,"Error in libraryread"
print *,"Resize array to capture all"
print *,"of library file"
stop
end if

print *,"Nuclides in library=",nlibrarysize
print *

!allocate targetnuclide and productnuclide arrays
allocate(targetnucli(nlibrarysize),productnucli(nlibrarysize))
allocate(librarynumbers(nlibrarynumberscategories,nlibrarysize))

```

```

allocate(converselibrarylocation(nlibrarysize))
if(nisotopes>=1) allocate(librarylocation(nisotopes))

!read nuclides from library
do i=1,nlibrarysize
!file column mappings
!1=Product Nuclide
!2=Target Nuclide
!3=Thermal Cross section
!4=thermal x-s uncert
!5=fast x-s
!6=fast x-s uncert
!7=half life
!8=half life unit
!9=half life uncert
!10=half life uncert unit
!11=atomic mass
!12=atomic mass uncert
!13=Atomic Weight of element
!14=Atomic Weight uncert
!15=abundance
!16=abundance uncert
read(outputarray(i+offset,1),*) productnucli(i)
read(outputarray(i+offset,2),*) targetnucli(i)
!librarynumbers mapping
!1=thermal cross section

```

```

!2=thermal cross section uncert
!3=fast cross section
!4=fast cross section uncert
!5=half life
!6=half life uncert
!7=atomic mass
!8=atomic mass uncert
!9=atomic weight
!10=atomic weight uncert
!11=abundance
!12=abundance uncert
read(outputarray(i+offset,3),*) tempreal1 !Thermal cross
section
librarynumbers(1,i)=tempreal1*1d-24 !convert from barn to
cm^2
read(outputarray(i+offset,4),*) tempreal2 !thermal cross
section uncertainty
librarynumbers(2,i)=tempreal2*1d-24 !convert from barn to
cm^2
read(outputarray(i+offset,5),*) tempreal3 !fast cross
section
librarynumbers(3,i)=tempreal3*1d-24 !convert from barn to
cm^2
read(outputarray(i+offset,6),*) tempreal4 !fast cross
section uncertainty
librarynumbers(4,i)=tempreal4*1d-24 !convert from barn to

```

cm²

```
!Convert half life value to seconds
!timeconvert(half life number,unitin,unitout)
read(outputarray(i+offset,7),*) tempreal5 !half life number
tempreal5=timeconvert(tempreal5,&
outputarray(i+offset,8),"sec")
!convert from half life to lambda
if(tempreal5/=0d0) then
librarynumbers(5,i)=dlog(2.0d0)/tempreal5
else
librarynumbers(5,i)=huge(0d0)
end if

read(outputarray(i+offset,9),*) tempreal6 !half life
uncertainty
tempreal6=timeconvert(tempreal6,&
outputarray(i+offset,10),"sec")
!convert from half life to lambda
librarynumbers(6,i)=librarynumbers(5,i)*tempreal6/tempreal5

read(outputarray(i+offset,11),*) tempreal7 !atomic mass
librarynumbers(7,i)=tempreal7
read(outputarray(i+offset,12),*) tempreal8 !atomic mass
uncertainty
librarynumbers(8,i)=tempreal8
```

```

read(outputarray(i+offset,13),*) tempreal9 !atomic weight
librarynumbers(9,i)=tempreal9
read(outputarray(i+offset,14),*) tempreal10 !atomic weight
uncertainty
librarynumbers(10,i)=tempreal10
read(outputarray(i+offset,15),*) tempreal11 !abundance
librarynumbers(11,i)=tempreal11/1d2
read(outputarray(i+offset,16),*) tempreal12 !abundance
uncertainty
librarynumbers(12,i)=tempreal12/1d2
end do

print *,"Done reading library file"
print *

!trim character strings
do i=1,nlibrarysize
productnucli(i)=trim(adjustl(productnucli(i)))
targetnucli(i)=trim(adjustl(targetnucli(i)))
end do

print *,"Checking for duplicates in library"
print *

!check to make sure that library has no duplicates
if(nlibrarysize>=2) then

```

```

do i=1,nlibrarysize-1
do j=i+1,nlibrarysize
if(productnucli(i)==productnucli(j)) then
print *, "There are duplicate entries for the
same nuclide in the library"
print *, "Please remove one of the entries for
the following nuclide(s)"
print *, productnucli(i)
print *, "This way the program will know which
activation path to follow"
stop
end if
end do
end do
end if

print *, "Duplicate check completed in library"
print *

!return if there are no input isotopes to match against
if(nisotopes==0) return

print *, "Matching input entries to library"

!initialize librarylocation to 0
librarylocation(:)=0

```

```

converselibrarylocation(:)=0

!Assign locations of isotopes(:) to librarylocation(:)
do i=1,nisotopes
do j=1,nlibrarysize
if((isotopes(i)==productnucli(j)).or.&
(isotopes(i)==targetnucli(j))) then
librarylocation(i)=j !matching isotopes to the
    library
converselibrarylocation(j)=i !matching the library
to the isotopes
exit
end if
end do
end do

!make sure that all input nuclides are located in the library
do i=1,nisotopes
if(librarylocation(i)==0) then
print *, "Nuclide"
print *, isotopes(i)
print *, "not located in library"
print *, "Remove from input file or add to library"
stop
end if
end do

```



```

print *, "Input entries matched to library"
deallocate(outputarray)

return

end subroutine libraryread

!enter activation data directly in terminal
subroutine manualentry(outputfilenumber, isotopes, nisotopes,
activityinfo, activityuncertainty, samplesize)
use mody
implicit none

!imported variables
integer, intent(in) :: outputfilenumber !filenumber of output file
real(8), intent(inout) :: samplesize !mass of whole sample
character(*), allocatable, intent(inout) :: isotopes(:) !Names
of isotopes read from file
integer, intent(inout) :: nisotopes !Number of isotopes read
from file
real(8), allocatable, intent(inout) :: activityinfo(:),
activityuncertainty(:) !activity for each isotope

!subroutine specific variables
integer :: tempint1
character(80) :: tempchar1, tempchar2
character(80) :: choice
integer :: i, j

```

```

real(8) :: tempreal1,tempreal2,tempreal3
integer :: nsubiso !maximum number of gamma lines for any isotope
integer, allocatable :: gammalines(:) !Array of number of
gammalines for each nuclide
real(8), allocatable :: nuclideinfo(:, :, :) !Array storing
information read from terminal

do
  !Enter sample mass
  print *, "Enter sample mass with a space and then mass unit"
  print *, "acceptable mass units"
  print *, "mt, kg, g, mg, ug, ng, pg, fg, lb, oz"
  read *, tempchar1,tempchar2
  read(tempchar1,*) tempreal1
  samplesize=massconvert(tempreal1,tempchar2,"g") !convert
  to grams

  print *, "Sample mass in grams"
  print *, samplesize

  !enter number of isotopes
  print *, "enter number of radioisotopes"
  read *, tempchar1
  read(tempchar1,*) nisotopes

  print *, "what is the maximum number of gamma lines for any

```

```

isotope"
read *,tempchar1
read (tempchar1,*) nsubiso

!allocate arrays
if(.not.allocated(isotopes)) allocate(isotopes(nisotopes))
if(.not.allocated(activityinfo)) allocate(activityinfo
(nisotopes))
if(.not.allocated(activityuncertainty)) allocate
(activityuncertainty(nisotopes))
if(.not.allocated(gammalines)) allocate(gammalines
(nisotopes))
if(.not.allocated(nuclideinfo)) allocate(nuclideinfo
(nisotopes,nsubiso,5))

!set default variables
do i=1,nisotopes
do j=1,nsubiso
nuclideinfo(i,j,1)=1d0 !set id certainty to 1
end do
end do

print *,"Enter names of isotopes with all capital letters
and no space"
print *,"Examples"
print *,"CS-137"

```

```

print *, "TE-123m"
print *, "MN-56"
do i=1,nisotopes
print *, "Enter name of isotope",i
read *,tempchar1
isotopes(i)=tempchar1
print *, "number of gamma lines for isotope",i
read *,tempchar1
read(tempchar1,*) tempint1
gammalines(i)=tempint1
do j=1,tempint1
print *, "Enter energy of gamma (keV)",j
read *,tempchar1
read(tempchar1,*) tempreal1
nuclideinfo(i,j,2)=tempreal1
print *, "Enter Yield of gamma (%)",j
print *, "100% would be 100"
read *,tempchar1
read(tempchar1,*) tempreal1
nuclideinfo(i,j,3)=tempreal1
print *, "Enter Activity (Bq) at gamma energy",j
read *,tempchar1
read(tempchar1,*) tempreal1
nuclideinfo(i,j,4)=tempreal1
print *, "Energy Activity uncertainty for gamma",j
read *,tempchar1

```

```

read(tempchar1,*) tempreal1
nuclideinfo(i,j,5)=tempreal1
end do

end do

print *, "Summary"
print *, "Nuclides=", nisotopes
print *, "Printing isotopes and gamma lines"
do i=1,nisotopes
do j=1,gammalines(i)
isotopes(i)=trim(adjustl(isotopes(i)))
print *, isotopes(i),j
end do
end do

!check for correctness
print *, "Is this correct? (y/n)"
read *,tempchar1

select case(tempchar1)
case("y","Y","Yes","yes","YES")
!write to output file and exit loop
write(outputfilenumber,*) "Sample information
entered at terminal"
exit
case("n","N","No","no")

```

```

!go back and reenter all information
cycle
case default
!write to output file and exit loop
write(outputfilenumber,*) "Sample information
entered at terminal"
exit
end select
end do

!Calculate average activity and activity uncertainty
print *, "which activity average would you like?"
print *, ""
print *, "1-weighted mean"
print *, "2-arithmetic mean"
print *, "0-stop and exit"
read *, choice

do
select case(choice)
case("1")
print *, "taking weighted mean"
do i=1,nisotopes
!use weighted mean equation
!top=sum(x(i)/sigma(i)^2
!bottom=sum(1/sigma(i)^2)

```

```

tempreal1=0d0 !top of equation
tempreal2=0d0 !bottom of equation
tempreal3=0d0 !error equation
do j=1,gammalines(i)
tempreal1=tempreal1+(nuclideinfo(i,j,4)
/nuclideinfo(i,j,5)**2d0) !top
tempreal2=tempreal2+(1d0/nuclideinfo(i,j,5)
**2d0) !bottom
end do
activityinfo(i)=tempreal1/tempreal2
do j=1,gammalines(i)
tempreal3=tempreal3+(1d0/nuclideinfo(i,j,5)
**2d0)
end do
activityuncertainty(i)=dsqrt(1d0/tempreal3)
end do
exit
case("2")
print *,"taking arithmetic mean"
do i=1,nisotopes
tempreal1=0d0
tempreal2=0d0
do j=1,gammalines(i)
tempreal1=tempreal1+nuclideinfo(i,j,4)
!activity
tempreal2=tempreal2+nuclideinfo(i,j,5)

```

```

!activity uncertainty
end do
activityinfo(i)=tempreal1/dbl(e(gammalines(i))
do j=1,gammalines(i)
tempreal3=tempreal3+(1d0/nuclideinfo(i,j,5)
**2d0)
end do
activityuncertainty(i)=dsqrt(1d0/tempreal3)
end do
exit
case("0")
print *,"exiting"
stop
case default
print *,"select again"
cycle
end select
end do

!deallocate stuff
if(allocated(gammalines)) deallocate(gammalines)
if(allocated(nuclideinfo)) deallocate(nuclideinfo)

return
end subroutine

!this will calculate the barcode given a certain amount

```


!of material from each batch

!

!many subroutines used are not used for their original purpose.

Some variables

!are dummies just for the purpose of the subroutines even if they
aren't used.

```
subroutine massbarcode(inputfilenumber,outputfilenumber,  
batchfilenumber)
```

```
use mody
```

```
implicit none
```

```
integer,intent(in)::inputfilenumber !file to read oxides from
```

```
integer,intent(in)::batchfilenumber !file where batch information  
is stored
```

```
integer,intent(in)::outputfilenumber
```

```
!output file number where prescribed masses are output to
```

```
!specific variables
```

```
integer::i,j
```

```
integer::nbatches
```

```
!number of batches read from ratio text file
```

```
integer::nisotopes !number of isotopes in input file
```

```
character(500),allocatable::isotopes(:)
```

```
!stores the isotope names from the batch information file
```

```
real(8),allocatable::amatrix(:,:), xmatrix(:), bmatrix(:)
```

```
real(8),allocatable::nuclideinfo(:),measuredmass(:),
```

```

measuredmassfraction(:)
character(500),allocatable::tempisotopes(:)
real(8),allocatable::tempmeasuredmass(:)
character(80)::sampledescription
character(100)::tempchar1,tempchar2,tempchar3
real(8)::tempreal1
integer::tempint1,tempint2
integer::noxides,start,finish
logical::foundinoxidelist
character(1000)::barcodestring
real(8),allocatable::batchconcentrations(:,:)
!stores the concentration of batches of isotopes
character(1000),allocatable::batchnames(:,:)
!stores the names or id tags of the stock batch isotopes
real(8),allocatable::batchconcentrationerror(:,:)
character(1000),allocatable::inputarray(:,:)
integer,parameter::offset=1

!read sample description
print *,"Sample description"
print *,"If more than one word, put in quotes"
read *, sampledescription
print *

!put description in outputfile
print *,"Sample description=",sampledescription

```

```

print *
write(outputfilenumber,*) ' ';trim(adjustl
(sampledescription)),';'
write(outputfilenumber,*)

!read batch information
call batchread(batchfilenumber,2,batchconcentrations,&
batchconcentrationserror,batchnames,nbatches)

!Read measured masses file
print *, "Filename of measured masses in grams?"
call filenameassign(inputfilenumber)
call csvfilereader(inputarray,inputfilenumber,tempint1,
tempint2,";",'")')
close (inputfilenumber)
nisotopes=tempint1-offset

!make sure that measurements are fitted to the number
!of batches
if(nisotopes/=nbatches) then
print *, "Incorrect number of measurements"
print *, "to satisfy the number of batches used"
print *, "Number of measurements=",nisotopes
print *, "Number according to batches=",nbatches
stop
end if

```

```

!allocate arrays
if(.not.allocated(nuclideinfo)) allocate(nuclideinfo(nisotopes))
if(.not.allocated(isotopes)) allocate(isotopes(nisotopes))
if(.not.allocated(tempisotopes)) allocate(tempisotopes
(nisotopes))
if(.not.allocated(tempmeasuredmass)) allocate(tempmeasuredmass
(nisotopes))

!allocate matrix
if(.not.allocated(bmatrix)) allocate(bmatrix(nisotopes))
if(.not.allocated(measuredmass)) allocate(measuredmass
(nisotopes))
if(.not.allocated(measuredmassfraction)) allocate
(measuredmassfraction(nisotopes))

!initialize array
isotopes(:)=""

!read masses into array
do i=1,nisotopes
read(inputarray(i+offset,1),'(a)') tempchar1
tempisotopes(i)=trim(adjustl(tempchar1))
read(inputarray(i+offset,2),*) tempreal1
tempmeasuredmass(i)=tempreal1
end do

```

```

!check for duplicates
do i=1,nisotopes-1
do j=i+1,nisotopes
if(tempisotopes(i)==tempisotopes(j)) then
print *,"Error in massbarcode"
print *,"There are duplicates in the"
print *,"measured mass list"
stop
end if
end do
end do

!sort according to batch order and copy back to temp array
do i=1,nisotopes
do j=1,nbatches
if(batchnames(1,j)==tempisotopes(i)) then
isotopes(j)=tempisotopes(i)
measuredmass(j)=tempmeasuredmass(i)
end if
end do
end do

tempisotopes(:)=isotopes(:)
tempmeasuredmass(:)=measuredmass(:)

do i=1,nisotopes

```

```

if(isotopes(i)=="") then
print *,"Error in massbarcode"
print *,"There is a missing batch number"
print *,"from the list"
stop
end if
end do

print *,"Nuclides=",nisotopes
print *

print *,"Done reading input file"
print *

!read gravimetric list to find out weighted atomic masses for
each batch
print *,"Name of file containing a list of batches"
print *,"and their associated atomic weights and"
print *,"gravimetric information?"
print *,"column 1=batch name like CD-106 102123"
print *,"column 2=batch atomic weight"
print *,"column 3=gravimetric fraction by weight"
call filenameassign(inputfilenumber)
deallocate(inputarray)
call csvfilereader(inputarray,inputfilenumber,tempint1,tempint2
,"," ,'"')

```

```

close(inputfilenumber)

noxides=tempint1-offset

!search for matches
do i=1,nisotopes
foundinoxidelist=.false.
do j=1,noxides
tempchar1=trim(adjustl(inputarray(j+offset,1)))
tempchar2=trim(adjustl(inputarray(j+offset,2)))
tempchar3=trim(adjustl(inputarray(j+offset,3)))
if(tempchar1==batchnames(1,i)) then
foundinoxidelist=.true.
if(isnumeric(tempchar2)) then
read(tempchar2,*) tempreal1
bmatrix(i)=tempreal1
else
print *,"error in barcodemass"
print *,"expecting numerical value for atomic
weight"
print *,"in reading file for batch"
print *,tempchar1
stop
end if
if(isnumeric(tempchar3)) then
read(tempchar3,*) tempreal1

```

```

measuredmassfraction(i)=tempreal1/1d2
else
print *,"error in barcodemass"
print *,"expecting numerical value for
    gravimetric value"
print *,"in reading file for batch"
print *,tempchar1
stop
end if
exit
end if
end do

if(.not.foundinoxidelist) then
print *,"error in barcode mass"
print *,"a weighted atomic mass and gravimetric"
print *,"information were not found for batch"
print *,batchnames(1,i)
stop
end if
end do

!take into account gravimetric changes
tempmeasuredmass(:)=measuredmass(:)*measuredmassfraction(:)

!convert from weight% to atom%
call atommassconvert(2,tempmeasuredmass,bmatrix,nuclideinfo)

```



```

!output mass and atom% readings to output file
write(outputfilenumber,101) !mass header
do i=1,nbatches
write(outputfilenumber,102) trim(adjustl(batchnames(1,i))),&
measuredmass(i),nuclideinfo(i)
end do
write(outputfilenumber,*)

!translate to barcode
nuclideinfo(:)=nuclideinfo(:)*1d2
barcodestring=""
do i=1,nbatches
start=((i-1)*5)+1
finish=start+4
write(tempchar1,'(f5.0)') nuclideinfo(i)
read(tempchar1,*) tempreal1
tempint1=int(tempreal1)
write(tempchar1,'(i5.5)') tempint1
barcodestring(start:finish)=trim(adjustl(tempchar1))
end do

write(outputfilenumber,201) nbatches
write(outputfilenumber,202) trim(adjustl(barcodestring))

print 103,barcodestring

```

```

print *

!close output file
close(outputfilenumber)

!Deallocate all arrays
if(allocated(isotopes)) deallocate(isotopes)
if(allocated(tempisotopes)) deallocate(tempisotopes)
if(allocated(batchconcentrations)) deallocate
(batchconcentrations)
if(allocated(batchconcentrationserror)) then
deallocate(batchconcentrationserror)
end if
if(allocated(batchnames)) deallocate(batchnames)
if(allocated(nuclideinfo)) deallocate(nuclideinfo)
if(allocated(amatrix)) deallocate(amatrix)
if(allocated(xmatrix)) deallocate(xmatrix)
if(allocated(bmatrix)) deallocate(bmatrix)
if(allocated(measuredmass)) deallocate(measuredmass)
if(allocated(tempmeasuredmass)) deallocate(tempmeasuredmass)
deallocate(inputarray)

print *, "Program Complete"

101 format(';"Batch";"mass (g)";"atom %"')
102 format(';"',a20,'"';',f12.8,"";',f6.2,"";',f12.8)

```

```

103 format("Barcode= ",a100)
201 format(';;; "Number of Batches";',i10)
202 format(';;; "Barcode";"',a,'''')

return

end subroutine

subroutine matchbatch(requestedat,nlibrarysize,librarylocation,
converselibrarylocation,batchnames,&
nbatches,targetnucli,nisotopes)
implicit none
integer,allocatable, intent(inout) :: requestedat(:) !location
of batch isotopes in input file
integer, intent(in) :: nbatches !number of batches read from
ratio text file
integer, intent(in) :: nisotopes !number of isotopes from
input file
integer, intent(in) :: nlibrarysize !how big the library is
character(*), intent(in) :: batchnames(:, :)
!stores the names or id tags of the stock batch isotopes
character(*), intent(in) :: targetnucli(:) !stores the names
of isotopes in the library
integer, intent(in) :: librarylocation(:) !where in the library
input file isotopes are found
integer, intent(in) :: converselibrarylocation(:)
!where in the input file library entries are located

```

```

!specific variables
integer :: i,j,k

!allocate requestedat array
allocate(requestedat(nbatches))

!set default to the zero isotope if not in input file
requestedat=0

print *,"Matching isotopes in batch file to isotopes in library"
print *

!match batch isotopes to the input file
do i=1,nbatches
do j=1,nlibrarysize
if(batchnames(2,i)==targetnucli(j)) then
do k=1,nisotopes
if(librarylocation(k)==j) then
requestedat(i)=converselibrarylocation(j)
exit
end if
end do
end if
end do
end do

```

```

return

end subroutine

!this will evaluate the distance of the
!ampoules to one barcode
!that could match the calculated ampoule ratios


subroutine singlebarcodedistance(outputfilenumber,&
ampoules,ampouleserror,nbatches,batchnames)
use mody
implicit none
integer,intent(in)::outputfilenumber
real(8),intent(in)::ampoules(:)
real(8),intent(in)::ampouleserror(:)
integer,intent(in)::nbatches
character(*),intent(in)::batchnames(:, :)

!subroutine specific
integer::i,j
integer::barodelength,startpoint,endpoint
character(500)::barcode
real(8)::set1(nbatches),set2(nbatches)
real(8),allocatable::normarray(:)
integer,allocatable::barcodeintegers(:, :)
integer::nbarcodes
integer::tempint1
real(8)::tempreal1

```

```

!check number of batches
if(nbatches==1) then
print *,"I'm going to stop right now and declare that
since there is"
print *,"only 1 degree of freedom, there isn't a way to
generate"
print *,"different combinations of only 1 thing"
stop
end if

print *,"Enter barcode"
read *,barcode
barcode=trim(adjustl(barcode))

!make sure number of batches match
tempint1=len_trim(barcode)
barcodelength=tempint1/5
if(barcodelength/=nbatches) then
print *,"error in singlebarcodedistance"
print *,"barcode is incorrect length"
stop
end if

!dissect barcode into small pieces
do i=1,nbatches

```

```

startpoint=(i-1)*5+1
endpoint=startpoint+4
read(barcode(startpoint:endpoint),*) tempreal1
set2(i)=tempreal1/1d2
end do

nbarcodes=3 !itself,statistical error,actual error
allocate(normarray(nbarcodes))
allocate(barcodeintegers(nbarcodes,nbatches))

!calculate spectral distances
!distance between itself is 0
normarray(1)=0d0
!distance of statistical error
set1(:)=0d0
normarray(2)=barcodedistance(ampouleserror,set1)
!distance between 2 points
normarray(3)=barcodedistance(ampoules,set2)

!translate into barcodes
do i=1,nbatches
barcodeintegers(1,i)=int(ampoules(i)*1d2)
barcodeintegers(2,i)=int(ampouleserror(i)*1d2)
barcodeintegers(3,i)=int(set2(i)*1d2)
end do

```

```

!write batch heading information
write(outputfilenumber,*) ';"Barcode Section"'
write(outputfilenumber,'(";")',advance="no")
do i=1,nbatches
write(outputfilenumber,101,advance="no") batchnames(1,i)
end do
write(outputfilenumber,*) '"Whole Barcode";"Norm"'

!output barcodes into file
do i=1,nbarcodes
write(outputfilenumber,'(";")',advance="no")
if(i/=2) then
do j=1,nbatches
write(outputfilenumber,102,advance="no")
barcodeintegers(i,j)
end do

!all together
write(outputfilenumber,103,advance="no")
do j=1,nbatches
write(outputfilenumber,104,advance="no")
barcodeintegers(i,j)
end do
write(outputfilenumber,105,advance="no") !finish up
whole barcode
else

```



```

do j=1,nbatches
write(outputfilenumber,'(";")',advance="no")
end do
write(outputfilenumber,'(a)',advance="no") '"statistical
  error";'
end if

write(outputfilenumber,106) normarray(i)

end do

print *,"Original barcode norm, should be 0"
print *,normarray(1)
print *,"Statistical error norm"
print *,normarray(2)
print *,"Other barcode norm"
print *,normarray(3)
print *,"finished measuring distance between barcodes"
print *

deallocate(barcodeintegers)
deallocate(normarray)

101 format('"',a15,'";')
102 format('"',i5.5,'";')
103 format('"')

```

```

104 format(i5.5)
105 format('";')
106 format(f6.2)

end subroutine

!This will read in the isotopic makeup of standards against
!which the NAA sample can be compared to using the comparator
!method. This is more accurate than parametric NAA due to the
!cancellation of the flux and cross section terms in the equation.

subroutine standardreader(filenummer,nisotopes,nlibrarysize,
targetnucli,&
productnucli,librarynumbers,librarylocation,&
standardnumbers,standardavailable)
use mody
implicit none
integer,intent(in)::filenummer !unit number to use for reading
files
character(*),intent(in)::targetnucli(:),productnucli(:)
integer,intent(in)::nisotopes,nlibrarysize
real(8),allocatable,intent(out)::standardnumbers(:,:)
logical,allocatable,intent(out)::standardavailable(:)
real(8),intent(in)::librarynumbers(:,:)
integer,intent(in)::librarylocation(:)

!subroutine specific

```

```

real(8)::mass,massuncert
integer::nstandards,offset,listlength
integer::nlines
integer::tempint1
character(80)::tempchar1,tempchar2
real(8)::tempreal1,tempreal2,tempreal3,tempreal4
character(80),allocatable::tempnames(:)
character(200),allocatable::outputarray(:, :)
integer::categories=14
integer,allocatable::assignspot(:)
real(8),allocatable::tempnumbers(:, :)
real(8),allocatable::temparray1(:),temparray2(:),temparray3(:)
real(8),allocatable::temparray4(:),temparray5(:),temparray6(:)
integer::i,j,k

offset=5 !what line number the list starts on
offset=offset-1

print *, "How many standards are there?"
read *,tempchar1
read(tempchar1,*) nstandards

!allocate arrays
allocate(standardnumbers(nisotopes,categories))
allocate(standardavailable(nisotopes))

```

```

!initialize arrays
standardnumbers(:,:)=0d0
standardavailable(:)=.false.

if(nstandards==0) return

do i=1,nstandards
!Open file to read
print *, "Name of file with isotopic data"
print *, "about standard?"
call filenameassign(filenameumber)

!read file containing standard information
call csvfilereader(outputarray,filenameumber,nlines,tempint1,
";",',','')
close(filenameumber)

!count isotopes on list
listlength=nlines-offset

!allocate temp array
allocate(tempnumbers(listlength,categories))
allocate(tempnames(listlength))
allocate(assignspot(listlength))
tempnumbers(:,:)=0d0
tempnames(:)=""

```

```

assignspot(:)=0

!read mass
read(outputarray(1,2),*) tempreal1
read(outputarray(1,3),*) tempchar1
read(outputarray(2,2),*) tempreal2
read(outputarray(2,3),*) tempchar2

!convert mass to grams
mass=massconvert(tempreal1,tempchar1,"g")
massuncert=massconvert(tempreal2,tempchar2,"g")

!record standard sample mass
tempnumbers(:,9)=mass
tempnumbers(:,10)=massuncert

!read name and atom% into temp array
do j=1,listlength
read(outputarray(j+offset,1),*) tempchar1
tempchar1=trim(adjustl(tempchar1))
tempnames(j)=tempchar1
read(outputarray(j+offset,2),*) tempchar1
tempchar1=trim(adjustl(tempchar1))
if((tempchar1=="Natural").or.&
(tempchar1=="natural").or.&
(tempchar1=="NATURAL").or.&

```

```

(tempchar1=="Nat").or.&
(tempchar1=="NAT").or.&
(tempchar1=="nat")) then
do k=1,nlibrarysize
if(tempnames(j)==targetnucli(k)) then
!matching the library to the isotopes
tempnumbers(j,1)=librarynumbers(11,k)*1d2
!Natural Abundance
tempnumbers(j,2)=librarynumbers(12,k)*1d2
!natural abundance uncert
exit
end if
end do
else
read(outputarray(j+offset,2),*) tempreal1 !atom%
read(outputarray(j+offset,3),*) tempreal2 !atom%
uncert
read(outputarray(j+offset,4),*) tempreal3 !Activity
read(outputarray(j+offset,5),*) tempreal4 !Activity
uncert
tempnumbers(j,1)=tempreal1
tempnumbers(j,2)=tempreal2
tempnumbers(j,13)=tempreal3
tempnumbers(j,14)=tempreal4
end if
end do

```

```

!look up atomic masses and weights from library
do j=1,listlength
do k=1,nisotopes
if((tempnames(j)==targetnucli(librarylocation(k))))
then
tempnames(j)=productnucli(librarylocation(k))
assignspot(j)=k
tempnumbers(j,5)=librarynumbers(7,
librarylocation(k))
!Atomic mass
tempnumbers(j,6)=librarynumbers(8,
librarylocation(k))
!Atomic mass uncert
tempnumbers(j,7)=librarynumbers(9,
librarylocation(k))
!Atomic weight
tempnumbers(j,8)=librarynumbers(10,
librarylocation(k))
!atomic weight uncert
exit
end if
end do
end do

!check atomic masses to make sure that no isotopes

```

```

!are missing from library
do j=1,listlength
if((tempnumbers(j,5)==0d0).or.&
(tempnumbers(j,6)==0d0)) then
print *,"Error in standardreader"
print *,"Missing data from array"
print *
print *,tempnumbers(1,:)
stop
end if
end do

!break things into single temporary vectors
!for atom% to weight% conversion
allocate(temparray1(listlength),temparray2(listlength))
allocate(temparray3(listlength),temparray4(listlength))
allocate(temparray5(listlength),temparray6(listlength))

temparray1(:)=tempnumbers(:,1) !atom%
temparray2(:)=tempnumbers(:,5) !atomic mass
temparray4(:)=tempnumbers(:,2) !atom% uncertainty
temparray5(:)=tempnumbers(:,6) !atomic mass uncert

!calculate weight% and isotope mass in standard
call atommassconvert(1,temparray1,temparray2,&
temparray3,temparray4,temparray5,temparray6)

```



```

!copy answers
tempnumbers(:,3)=temparray3(:) !weight%
tempnumbers(:,4)=temparray6(:) !weight% uncert
deallocate(temparray1,temparray2,temparray3)
deallocate(temparray4,temparray5,temparray6)

!calculate mass of isotope in standard
!mass=(weight fraction)*(whole standard mass)
tempnumbers(:,11)=tempnumbers(:,3)/1d2*tempnumbers(:,9)

do j=1,listlength
  if(tempnumbers(j,3)>0d0) then
    tempnumbers(j,12)=tempnumbers(j,11)*&
    dsqrt((tempnumbers(j,4)/tempnumbers(j,3))*2d0+&
    (tempnumbers(j,10)/tempnumbers(j,9))*2d0)
  else
    tempnumbers(j,12)=0d0
  end if
end do

!copy tempnumbers to standardnumbers
do j=1,listlength
  if(assignspot(j)>0) then
    standardnumbers(assignspot(j),:)=tempnumbers(j,:)
    standardavailable(assignspot(j))=.true.
  end if
end do

```

```

end if
end do

deallocate(tempnumbers,tempnames)
deallocate(assignspot)
end do

deallocate(outputarray)

end subroutine

!this will calculate the distance between two barcodes

subroutine twobarcodedistance()
use mody
implicit none

!subroutine specific
real(8)::tempreal1,tempreal2
real(8)::finaldistance
real(8),allocatable::set1(:),set2(:)
character(200)::barcode1,barcode2
integer::nbatches
integer::i,j
integer::startpoint,endpoint

!obtain barcodes

```

```

print *, "Please enter barcode 1 with no spaces"
read *, barcode1
print *
print *, "Please enter barcode 2 with no spaces"
read *, barcode2

!determine number of batches and make sure they match
i=len_trim(barcode1)
j=len_trim(barcode2)
if(i/=j) then
print *, "error in twobarcodedistance"
print *, "The lengths of the barcodes must be equal"
stop
end if

j=i/5*5
if(i/=j) then
print *, "error in twobarcodedistance"
print *, "the barcode length must be a multiple of 5"
stop
else
nbatches=i/5
end if

allocate(set1(nbatches), set2(nbatches))

```

```

!dissect barcode into small pieces
do i=1,nbatches
startpoint=(i-1)*5+1
endpoint=startpoint+4
read(barcode1(startpoint:endpoint),*) tempreal1
read(barcode2(startpoint:endpoint),*) tempreal2
set1(i)=tempreal1/1d2
set2(i)=tempreal2/1d2
end do

finaldistance=barcodedistance(set1,set2)

print *,"The residual between the two barcodes is"
print *,finaldistance
print *

deallocate(set1,set2)
end subroutine

```

APPENDIX B

GENIE-2000 INPUT FILES

Sample Genie-2000 Input File for a Solid Phase Taggant

```
*****
*****      G A M M A    S P E C T R U M    A N A L Y S I S
*****
```

Filename: C:\zak 103 folder\cdte 1\cdte 1 count 1.CNF

Report Generated On : 8/29/2011 9:12:11 AM

Sample Title : CdTe 1, count 1, 48cm

Sample Description : CdTe 1, count 1, 48cm

Sample Identification : CdTe 1

Sample Type :

Sample Geometry : 48cm

Peak Locate Threshold : 5.00

Peak Locate Range (in channels) : 1 - 65535

Peak Area Range (in channels) : 1 - 65535

Identification Energy Tolerance : 3.000 keV

Sample Size : 1.000E+000 Unit

Sample Taken On : 8/22/2011 10:08:50 AM

Acquisition Started : 8/23/2011 4:04:08 PM

Live Time : 3600.0 seconds

Real Time : 3707.5 seconds

Dead Time : 2.90 %

Energy Calibration Used Done On : 8/29/2011

Efficiency Calibration Used Done On : 8/29/2011

Efficiency ID : room 103, 48cm

Peak Analysis Report 8/29/2011 9:12:12 AM

***** P E A K A N A L Y S I S R E P O R T

Detector Name: DET1

Sample Title: CdTe 1, count 1, 48cm

Peak Analysis Performed on: 8/29/2011 9:12:12 AM

Peak Analysis From Channel: 1

Peak Analysis To Channel: 4096

Peak No.	ROI start	ROI end	Peak centroid	Energy (keV)	FWHM (keV)	Net Peak Area	Net Area Uncert.
1	38-	45	41.00	20.11	1.27	3.63E+005	1284.80
2	117-	126	123.00	61.12	0.67	7.96E+003	1786.45
3	140-	154	150.66	74.95	1.07	1.11E+005	2546.97
4	181-	190	187.00	93.12	0.78	2.37E+006	2407.89
5	213-	223	219.18	109.21	0.89	1.41E+004	1987.25
6	262-	272	269.00	134.12	0.75	1.41E+004	2060.79
7	311-	325	318.62	158.93	1.04	6.60E+005	3059.40
8	404-	413	406.37	202.81	0.98	9.51E+003	1686.47
9	428-	438	430.99	215.12	0.84	1.13E+004	1718.67
10	459-	470	463.50	231.38	1.13	3.50E+005	1849.81
11	518-	530	522.44	260.85	1.10	7.97E+005	1882.69
12	532-	542	534.51	266.88	1.12	3.94E+004	1383.06
13	567-	572	569.38	284.32	0.94	2.91E+003	853.94
M 14	642-	665	649.87	324.57	3.02	5.62E+004	990.56
m 15	642-	665	656.36	327.81	3.02	4.60E+004	1023.62
16	665-	681	673.32	336.29	1.09	1.61E+007	4377.81
M 17	719-	738	721.40	360.34	1.02	1.04E+004	335.25
m 18	719-	738	729.61	364.44	1.02	4.50E+004	411.09
19	832-	842	836.59	417.93	1.17	8.72E+004	597.89
20	843-	855	846.85	423.06	0.99	2.83E+003	574.70
21	910-	922	919.54	459.41	1.25	2.08E+003	536.50

22	955-	963	959.68	479.48	1.23	1.35E+004	412.84
23	978-	991	985.30	492.29	1.23	1.78E+006	1468.39
24	1013-	1032	1022.69	510.99	2.87	1.25E+005	755.76
25	1046-	1066	1056.32	527.81	1.26	5.76E+006	2451.11
26	1096-	1113	1103.65	551.47	1.67	5.67E+003	243.74
27	1187-	1198	1195.27	597.28	1.65	1.55E+003	157.99
28	1227-	1244	1237.66	618.48	1.70	4.48E+003	215.64
29	1247-	1257	1250.81	625.05	1.52	8.41E+002	141.85
30	1269-	1280	1274.79	637.05	1.34	2.13E+003	150.48
31	1335-	1349	1345.57	672.44	1.35	2.01E+003	171.17
32	1368-	1378	1372.13	685.72	1.39	1.10E+004	162.85
33	1387-	1401	1392.55	695.93	1.17	1.20E+003	158.05
M 34	1536-	1559	1547.19	773.26	1.72	3.24E+003	75.94
m 35	1536-	1559	1553.53	776.43	1.72	1.76E+003	66.10
36	1582-	1604	1593.72	796.52	1.51	7.17E+003	215.55
37	1627-	1641	1632.13	815.73	1.45	2.05E+003	145.27
38	1650-	1664	1658.57	828.95	1.53	1.61E+004	188.82
39	1700-	1709	1705.12	852.22	1.21	3.10E+002	104.78
M 40	1722-	1741	1728.46	863.89	1.54	1.61E+003	66.45
m 41	1722-	1741	1736.26	867.79	1.54	5.18E+002	52.82
42	1791-	1803	1796.89	898.11	1.49	1.21E+003	118.82

Peak Analysis Report

8/29/2011

9:12:12 AM

Peak No.	ROI start	ROI end	Peak centroid	Energy (keV)	FWHM (keV)	Net Peak Area	Net Area Uncert.
-------------	--------------	------------	------------------	-----------------	---------------	------------------	---------------------

43	1845-	1855	1851.07	925.20	1.48	2.86E+002	104.92
44	1865-	1873	1868.07	933.70	1.56	1.63E+003	96.56
45	2081-	2099	2088.77	1044.06	1.32	3.71E+002	175.26
46	2257-	2267	2260.63	1130.00	1.27	4.53E+002	132.29
47	2571-	2588	2581.68	1290.53	1.87	5.57E+002	101.50
48	2621-	2642	2635.34	1317.36	1.34	3.89E+002	114.83
49	2726-	2751	2737.65	1368.52	1.86	1.07E+005	349.42
50	3042-	3063	3050.06	1524.74	1.53	8.29E+002	93.64
51	3178-	3202	3192.69	1596.05	1.89	4.55E+003	119.33
52	3454-	3479	3464.33	1731.88	2.14	9.83E+003	151.83

M = First peak in a multiplet region

m = Other peak in a multiplet region

F = Fitted singlet

Errors quoted at 1.000 sigma

Interference Corrected Activity Report 8/29/2011 9:12:13 AM

 ***** N U C L I D E I D E N T I F I C A T I O N R E P O R T *****

Sample Title: CdTe 1, count 1, 48cm

Nuclide Library Used: C:\zak 103 folder\zak 08262011.nlb

..... IDENTIFIED NUCLIDES

Nuclide	Id	Energy	Yield	Activity	Activity
Name	Confidence	(keV)	(%)	(Bq /Unit)	Uncertainty
NA-22	0.587	511.00*	181.00	8.39967E+003	2.83481E+002
		1274.55	99.90		
NA-24	0.980	1368.55*	100.00	2.01119E+005	9.38973E+003
		2754.05	99.94		
K-42	0.971	1524.58*	18.80	1.31772E+004	1.85762E+003
SE-75	0.480	121.11	16.70	1.05028E+003	1.81531E+002
		136.00*	59.20		
		264.65*	59.80		
		279.53	25.20		
		400.65	11.40		
GE-77	0.538	211.03	30.80	2.26043E+004	4.02668E+003
		215.50*	28.60		
		264.44*	53.90		
		416.33*	21.80		
BR-82x	0.894	554.35*	70.80	2.23747E+003	1.24939E+002
		619.11*	43.40	3.25349E+003	1.95930E+002
		698.37*	28.50	1.50683E+003	2.05588E+002
		776.52*	83.50	8.42214E+002	4.48619E+001
		827.83*	24.00	2.86697E+004	1.14562E+003
		1044.00*	27.20	7.34300E+002	3.47576E+002
		1317.47*	26.50	9.80268E+002	2.92556E+002

		1474.88	16.32		
Y-90m	0.644	202.51*	96.58	1.36634E+006	2.73577E+005
		479.53*	90.71	4.38874E+006	2.21920E+005
CD-107	0.899	93.12*	4.70	1.12585E+008	5.64668E+006
		511.00*	0.40	2.03849E+008	7.17679E+006
		796.46*	0.06	1.14444E+008	5.38432E+006
		828.93*	0.16	1.06559E+008	4.01450E+006
CD-115	0.998	260.90*	1.94	4.35762E+006	3.36131E+005
		336.24*	45.90	4.64219E+006	2.82752E+005
		527.90*	27.40	4.35020E+006	1.71760E+005
CD-115m	1.000	933.84*	2.00	1.89766E+004	6.76306E+003
TE-123m	0.998	88.46	0.09		
		158.99*	84.00	3.69874E+004	3.38102E+003
TE-127	0.950	360.30*	0.14	1.08425E+007	6.82321E+005
		417.90*	0.99	1.43117E+007	6.40703E+005
TE-129m	0.572	105.50	0.14		
		556.56	0.12		
		695.84*	2.99	6.98217E+003	9.47574E+002
		729.53	0.70		
I-131	1.000	364.48*	81.20	5.51205E+003	2.93911E+002
TE-131m	0.481	773.68*	37.70	3.89658E+003	2.48141E+002
Interference Corrected Activity Report				8/29/2011	9:12:13 AM

Nuclide	Id	Energy	Yield	Activity	Activity
Name	Confidence	(keV)	(%)	(Bq /Unit)	Uncertainty

TE-131m	0.481	782.48	8.06		
		793.76*	13.10	2.56373E+004	1.69970E+003
		822.78	6.38		
		852.24*	19.20	8.10493E+002	2.77884E+002
		1125.48	11.20		
		1206.59	9.63		
LA-140	0.491	328.76*	20.60	3.39914E+004	2.24206E+003
		487.02	44.30		
		815.77*	22.90	3.43138E+003	2.76984E+002
		1596.21*	95.40	3.41237E+003	2.91714E+002
ND-147	0.613	91.11*	27.90	3.91106E+005	2.46411E+004
		531.02	13.10		
DY-157	0.858	182.20	1.84		
		265.34*	0.19	3.34259E+007	9.23563E+006
		326.16*	92.00	1.16898E+005	9.01097E+003
GD-159	0.974	363.56*	11.00	1.47501E+005	4.09378E+004
W-187	0.992	479.55*	21.80	2.15761E+004	1.27365E+003
		685.73*	27.30	2.03277E+004	1.06831E+003
IR-194	0.981	328.45*	13.10	1.09964E+005	1.66248E+004
PT-197	0.646	77.35*	17.00	1.20091E+005	2.00813E+004
		191.44	3.70		

* = Energy line found in the spectrum.

@ = Energy line not used for Weighted Mean Activity

Energy Tolerance : 3.000 keV

Nuclide confidence index threshold = 0.30

Errors quoted at 1.000 sigma

Interference Corrected Activity Report 8/29/2011 9:12:13 AM

***** I N T E R F E R E N C E C O R R E C T E D R E P O R T

	Nuclide		Wt mean	Wt mean
Nuclide	Id		Activity	Activity
Name	Confidence		(Bq /Unit)	Uncertainty
NA-22	0.587		4.202435E+003	3.113794E+002
NA-24	0.980		2.011191E+005	9.389733E+003
K-42	0.971		1.317719E+004	1.857619E+003
X CU-64	0.961			
SE-75	0.480		1.172275E+003	1.661464E+002
GE-77	0.538		2.982020E+004	2.962709E+003
BR-82x	0.894		1.091794E+003	3.915980E+001
Y-90m	0.644		7.750871E+005	1.869371E+005
CD-107	0.899		1.018613E+008	3.288942E+006
CD-115	0.998		4.417475E+006	1.345505E+005
CD-115m	1.000		1.897658E+004	6.763061E+003

	TE-123m	0.998	3.698738E+004	3.381016E+003
	TE-127	0.950	1.217425E+007	4.898212E+005
	TE-129m	0.572	1.923138E+003	9.647892E+002
?	I-131	1.000	5.512050E+003	2.939111E+002
	TE-131m	0.481	3.083408E+003	2.034120E+002
	LA-140	0.491	3.422595E+003	1.983139E+002
	ND-147	0.613	3.725258E+004	2.207634E+004
	DY-157	0.858	1.169257E+005	7.440855E+003
?	GD-159	0.974	1.475010E+005	4.093775E+004
X	DY-165	0.338		
X	HF-181	0.912		
	W-187	0.992	1.937810E+004	6.965141E+002
	IR-194	0.981	9.889155E+004	6.963496E+003
X	HG-197	0.474		
	PT-197	0.646	1.200910E+005	2.008133E+004

? = nuclide is part of an undetermined solution

X = nuclide rejected by the interference analysis

@ = nuclide contains energy lines not used in Weighted Mean Activity

Errors quoted at 1.000 sigma

Interference Corrected Activity Report 8/29/2011 9:12:13 AM

***** U N I D E N T I F I E D P E A K S *****

Peak Locate Performed on: 8/29/2011 9:12:12 AM

Peak Locate From Channel: 1

Peak Locate To Channel: 4096

Peak No.	Energy (keV)	Peak Size in Counts per Second	Peak CPS % Uncertainty	Peak Type
1	20.11	5.9301E+000	0.35	D-Esc.
2	61.12	1.3002E-001	22.45	Tol.
5	109.21	2.3054E-001	14.08	Tol.
10	231.38	5.7228E+000	0.53	Sum
13	284.32	4.7613E-002	29.31	Tol.
20	423.06	4.6245E-002	20.31	Sum
21	459.41	3.4047E-002	25.75	Sum
23	492.29	2.9072E+001	0.08	Sum
27	597.28	2.5385E-002	10.17	Sum
29	625.05	1.3744E-002	16.86	Sum
30	637.05	3.4765E-002	7.07	Sum
31	672.44	3.2789E-002	8.53	Sum
M 40	863.89	2.6369E-002	4.12	Sum
m 41	867.79	8.4628E-003	10.20	Sum
42	898.11	1.9747E-002	9.83	Sum
43	925.20	4.6791E-003	36.64	Sum
46	1130.00	7.4012E-003	29.21	Sum

47	1290.53	9.1017E-003	18.22	
52	1731.88	1.6063E-001	1.54	Sum

M = First peak in a multiplet region

m = Other peak in a multiplet region

F = Fitted singlet

Errors quoted at 1.000 sigma

Sample Genie-2000 Input File for a Liquid Phase Taggant

```
*****
*****      G A M M A    S P E C T R U M    A N A L Y S I S
*****
```

Filename: C:\GENIE2K\CAMFILES\ZAK KULAGE\ZAK-1 C1.CNF

Report Generated On : 10/26/2011 8:55:05 AM

Sample Title : ZAK 1 C1

Spectrum Description :

Sample Identification : ZAK 1

Sample Type : AMPOULE

Sample Geometry :

Peak Locate Threshold : 3.00

Peak Locate Range (in channels) : 1 - 16384
Peak Area Range (in channels) : 1 - 16384
Identification Energy Tolerance : 1.500 keV

Sample Size : 1.000E+000

Sample Taken On : 10/18/2011 12:16:35 PM

Acquisition Started : 10/19/2011 2:27:56 PM

Live Time : 3600.0 seconds

Real Time : 3855.4 seconds

Dead Time : 6.63 %

Energy Calibration Used Done On : 10/26/2011

Efficiency Calibration Used Done On : 10/26/2011

Efficiency ID : DUCK 48 CM

Peak Analysis Report 10/26/2011 8:55:05 AM

***** P E A K A N A L Y S I S R E P O R T

Detector Name: DUCK

Sample Title: ZAK 1 C1

Peak Analysis Performed on: 10/26/2011 8:55:04 AM

Peak Analysis From Channel: 1

Peak Analysis To Channel: 16384

	Peak No.	ROI start	ROI end	Peak centroid	Energy (keV)	FWHM (keV)	Net Peak Area	Net Area Uncert.
	1	108-	117	113.31	27.50	0.63	8.53E+003	210.24
M	2	294-	315	299.80	72.99	1.10	3.22E+003	172.99
m	3	294-	315	309.16	75.27	1.11	5.34E+003	193.50
	4	324-	336	330.52	80.48	1.10	2.06E+004	379.81
M	5	343-	365	349.69	85.15	1.24	4.12E+003	200.44
m	6	343-	365	359.82	87.62	1.25	1.77E+003	176.08
	7	377-	389	383.62	93.43	1.00	2.71E+004	386.86
	8	414-	426	420.43	102.41	1.00	1.30E+004	370.27
	9	546-	560	553.56	134.88	1.56	2.80E+003	418.02
	10	608-	623	615.94	150.10	1.01	3.90E+004	456.31
	11	647-	661	654.06	159.40	1.07	2.68E+004	423.51
	12	721-	736	728.80	177.63	0.94	2.84E+003	410.32
	13	742-	755	749.42	182.66	1.22	3.24E+003	368.45
	14	777-	785	781.19	190.41	0.54	6.82E+002	257.21
	15	817-	832	824.90	201.07	1.06	1.22E+004	406.25
	16	942-	959	951.06	231.85	1.14	5.50E+003	405.29

	17	981-	998	990.17	241.39	1.20	1.09E+004	408.86
M	18	1036-	1054	1039.39	253.39	0.29	3.68E+002	47.95
m	19	1036-	1054	1048.16	255.53	0.29	3.16E+002	54.71
	20	1063-	1080	1071.98	261.34	1.19	1.22E+004	396.04
	21	1092-	1103	1097.05	267.46	0.76	2.44E+002	263.78
	22	1136-	1152	1143.87	278.88	1.26	2.56E+003	330.47
	23	1159-	1177	1168.08	284.78	1.18	4.21E+004	382.64
	24	1268-	1281	1272.42	310.23	0.78	5.36E+002	242.02
	25	1332-	1346	1337.96	326.22	1.23	1.04E+003	257.00
	26	1371-	1390	1381.17	336.76	1.22	2.71E+005	616.46
	27	1402-	1415	1408.48	343.42	0.91	7.00E+002	227.85
	28	1441-	1449	1444.54	352.22	0.42	2.21E+002	154.66
	29	1486-	1506	1496.74	364.95	1.25	4.56E+005	733.27
	30	1526-	1536	1530.15	373.10	0.62	1.32E+001	149.31
	31	1572-	1581	1576.86	384.50	0.98	3.80E+002	135.21
M	32	1685-	1726	1690.89	412.31	1.38	9.81E+002	96.50
m	33	1685-	1726	1716.09	418.46	1.39	3.62E+003	122.18
	34	1770-	1786	1775.93	433.06	0.93	4.25E+002	188.16
	35	1846-	1866	1857.25	452.89	1.30	5.22E+003	227.65
M	36	1879-	1933	1886.59	460.05	1.28	3.19E+003	110.11
m	37	1879-	1933	1900.77	463.51	1.28	1.63E+003	91.83
m	38	1879-	1933	1923.51	469.06	1.29	3.85E+002	81.20
	39	1959-	1978	1968.64	480.06	1.27	1.98E+003	190.99
M	40	1991-	2031	2000.15	487.75	1.38	8.93E+002	85.55
m	41	1991-	2031	2020.84	492.80	1.38	3.42E+004	209.62
	42	2055-	2076	2064.82	503.53	1.29	8.76E+002	202.23

Peak Analysis Report

10/26/2011 8:55:05 AM

	Peak No.	ROI start	ROI end	Peak centroid	Energy (keV)	FWHM (keV)	Net Peak Area	Net Area Uncert.
	43	2086-	2108	2097.52	511.50	2.56	3.70E+004	293.30
	44	2155-	2177	2166.48	528.32	1.42	1.06E+005	381.84
M	45	2255-	2285	2262.35	551.71	1.49	2.94E+002	78.53
m	46	2255-	2285	2275.42	554.90	1.50	4.79E+002	86.34
	47	2394-	2418	2406.24	586.81	1.38	1.41E+003	193.66
	48	2462-	2478	2471.06	602.62	1.29	8.22E+002	136.86
	49	2528-	2548	2539.38	619.28	0.82	7.97E+002	155.23
M	50	2601-	2649	2614.09	637.51	1.51	2.54E+004	178.48
m	51	2601-	2649	2637.73	643.27	1.52	9.30E+002	70.59
	52	2719-	2740	2729.07	665.55	1.54	2.75E+003	156.71
	53	2802-	2823	2814.20	686.32	1.57	1.34E+003	148.04
	54	2848-	2868	2855.35	696.36	1.44	1.82E+003	143.30
	55	2917-	2933	2926.52	713.72	1.70	9.55E+002	115.62
M	56	2955-	3001	2966.21	723.40	1.58	5.57E+003	103.26
m	57	2955-	3001	2992.76	729.87	1.59	3.63E+002	60.82
	58	3041-	3066	3053.65	744.73	1.00	7.89E+002	148.61
	59	3163-	3188	3174.69	774.25	1.62	2.36E+004	211.40
	60	3197-	3224	3210.82	783.07	1.54	4.35E+003	165.08
	61	3243-	3270	3257.14	794.37	1.62	8.24E+003	175.16
	62	3362-	3385	3376.33	823.44	1.57	3.44E+003	143.15

M	63	3482-	3529	3496.84	852.83	1.73	1.21E+004	129.39
m	64	3482-	3529	3514.82	857.22	1.74	8.78E+002	62.74
	65	3723-	3746	3733.91	910.66	1.81	1.83E+003	133.56
	66	3769-	3787	3778.05	921.43	0.96	4.86E+002	110.51
M	67	3852-	3898	3862.45	942.02	0.89	1.39E+002	143.85
m	68	3852-	3898	3893.11	949.50	0.90	8.58E+001	88.69
	69	4333-	4363	4348.46	1060.57	1.44	1.10E+003	167.63
	70	4396-	4410	4402.96	1073.86	0.55	1.27E+002	94.64
	71	4603-	4634	4618.91	1126.54	1.86	5.57E+003	200.38
	72	4698-	4729	4713.80	1149.69	3.01	2.22E+003	184.73
	73	4936-	4965	4952.50	1207.91	1.94	4.20E+003	145.49
	74	5069-	5091	5079.12	1238.80	1.00	2.10E+002	92.64
	75	5189-	5211	5198.46	1267.91	0.81	1.21E+002	77.41
	76	5387-	5411	5399.10	1316.85	0.33	3.54E+002	78.79
	77	5601-	5635	5618.17	1370.29	2.16	1.59E+005	411.41
	78	6536-	6562	6554.45	1598.67	1.41	3.85E+002	67.64
	79	6745-	6772	6758.58	1648.47	1.28	4.27E+002	69.49
	80	7092-	7129	7110.96	1734.42	2.96	9.22E+003	133.16
	81	7742-	7767	7753.41	1891.14	0.85	2.90E+002	71.31
	82	8204-	8231	8218.45	2004.57	2.00	5.37E+002	79.80
	83	9087-	9106	9095.98	2218.63	0.27	5.36E+001	58.77
	84	9190-	9233	9211.96	2246.92	3.82	1.38E+004	165.10
	85	9463-	9480	9471.28	2310.17	0.66	1.54E+001	59.12
	86	11291-	11337	11314.16	2759.71	3.28	8.60E+004	298.49

M = First peak in a multiplet region

m = Other peak in a multiplet region

F = Fitted singlet

Errors quoted at 1.000 sigma

Interference Corrected Activity Report 10/26/2011 8:55:05 AM

***** N U C L I D E I D E N T I F I C A T I O N R E P O R T

Sample Title: ZAK 1 C1

Nuclide Library Used: C:\GENIE2K\CAMFILES\ZAK KULAGE\genie nuc

..... IDENTIFIED NUCLIDES

Nuclide	Id	Energy	Yield	Activity	Activity
Name	Confidence	(keV)	(%)	(Bq /)	Uncertainty
NA-22	0.547	511.00*	181.00	3.86830E+004	1.35762E+003
		1274.55	99.90		
KR-85	0.605	151.18*	75.10	2.84237E+006	2.73523E+005
		304.86	13.70		
Y-90m	0.655	202.51*	96.58	3.98511E+006	4.17178E+005
		479.53*	90.71	1.22728E+006	1.28416E+005
ZR-97	0.860	743.33*	94.80	6.41074E+003	1.22866E+003

RU-105	0.390	316.49	11.00		
		469.35*	17.30	2.43629E+005	5.22023E+004
		676.34	15.50		
		724.27*	46.70	1.88270E+006	7.58763E+004
CD-107	0.877	93.12*	4.70	9.05180E+006	4.89495E+005
		511.00*	0.40	2.93778E+008	1.06150E+007
		796.46	0.06		
		828.93	0.16		
CD-109	0.988	88.03*	3.61	4.70900E+004	5.47360E+003
IN-114m	0.566	190.28*	15.40	4.40703E+003	1.71851E+003
		558.43	4.38		
		725.24	4.33		
CD-115	0.982	260.90*	1.94	1.02031E+006	9.09646E+004
		336.24*	45.90	1.13291E+006	7.37933E+004
		527.90*	27.40	1.06068E+006	4.27329E+004
TE-123m	0.988	88.46*	0.09	1.85657E+006	2.09584E+005
		158.99*	84.00	2.97378E+004	2.94967E+003
SB-124	0.606	602.71*	97.87	1.84982E+003	3.14027E+002
		1691.02	47.10		
TE-127	0.749	360.30	0.14		
		417.90*	0.99	4.17344E+006	2.37659E+005
TE-129m	0.842	105.50	0.14		
		556.56	0.12		
		695.84*	2.99	1.52845E+005	1.31820E+004
		729.53*	0.70	1.36367E+005	2.33686E+004
I-131	0.984	364.48*	81.20	8.91873E+005	5.01716E+004

TE-131m	0.957	773.68*	37.70	3.10527E+005	1.68779E+004
		782.48*	8.06	2.70104E+005	1.77323E+004
		793.76*	13.10	3.18412E+005	1.83417E+004
		822.78*	6.38	2.81245E+005	1.90250E+004
		852.24*	19.20	3.38509E+005	1.83068E+004
		1125.48*	11.20	3.32613E+005	2.03905E+004
		1206.59*	9.63	3.06641E+005	1.89759E+004
SM-153	0.957	103.18*	28.30	6.25217E+004	4.04190E+003
DY-157	0.946	182.20*	1.84	1.61614E+006	3.01359E+005
Interference Corrected Activity Report				10/26/2011	8:55:05 AM

Nuclide Name	Id Confidence	Energy (keV)	Yield (%)	Activity (Bq /)	Activity Uncertainty
DY-157	0.946	265.34	0.19		
		326.16*	92.00	1.43205E+004	3.70725E+003
GD-159	0.863	363.56*	11.00	1.60804E+007	4.47210E+006
TM-170	0.950	84.30*	3.26	1.23301E+005	9.23022E+003
W-187	0.972	479.55*	21.80	3.52765E+004	3.86114E+003
		685.73*	27.30	2.58648E+004	3.13828E+003
HG-197	0.465	70.82	16.60		
		133.98*	34.00	1.60189E+004	3.35921E+003
AU-198	0.981	411.80*	95.58	2.16311E+003	2.33527E+002
HG-203	0.993	279.20*	81.46	3.82484E+003	5.76201E+002

* = Energy line found in the spectrum.

@ = Energy line not used for Weighted Mean Activity

Energy Tolerance : 1.500 keV

Nuclide confidence index threshold = 0.30

Errors quoted at 1.000 sigma

Interference Corrected Activity Report 10/26/2011 8:55:05 AM

***** I N T E R F E R E N C E C O R R E C T E D R E P O R T

	Nuclide		Wt mean	Wt mean
	Nuclide	Id	Activity	Activity
	Name	Confidence	(Bq /)	Uncertainty
	NA-22	0.547	3.749108E+004	1.359106E+003
X	CU-64	0.948		
	KR-85	0.605	2.842374E+006	2.735231E+005
	Y-90m	0.655	8.289313E+005	1.529053E+005
	ZR-97	0.860	6.410738E+003	1.228662E+003
	RU-105	0.390	7.791869E+005	4.310734E+004
	CD-107	0.877	9.051794E+006	4.894950E+005
	CD-109	0.988	4.633574E+004	5.316418E+003

X	PD-109	0.969		
X	PD-109x	0.970		
	IN-114m	0.566	4.407032E+003	1.718515E+003
	CD-115	0.982	1.070523E+006	3.426084E+004
	TE-123m	0.988	2.973782E+004	2.949670E+003
	SB-124	0.606	1.849825E+003	3.140272E+002
	TE-127	0.749	4.173439E+006	2.376591E+005
	TE-129m	0.842	1.488675E+005	1.148130E+004
?	I-131	0.984	8.918727E+005	5.017158E+004
	TE-131m	0.957	3.099364E+005	1.349366E+004
	SM-153	0.957	6.252166E+004	4.041898E+003
X	EU-155	0.301		
	DY-157	0.946	1.456327E+004	3.707083E+003
?	GD-159	0.863	1.608045E+007	4.472101E+006
	TM-170	0.950	1.233014E+005	9.230221E+003
X	LU-176M	0.744		
	W-187	0.972	2.004814E+004	2.907764E+003
	HG-197	0.465	1.601890E+004	3.359210E+003
	AU-198	0.981	2.163115E+003	2.335267E+002
	HG-203	0.993	3.824837E+003	5.762012E+002

? = nuclide is part of an undetermined solution

X = nuclide rejected by the interference analysis

@ = nuclide contains energy lines not used in Weighted Mean
Activity

Errors quoted at 1.000 sigma

Interference Corrected Activity Report 10/26/2011 8:55:05 AM

***** U N I D E N T I F I E D P E A K S *****

Peak Locate Performed on: 10/26/2011 8:55:04 AM

Peak Locate From Channel: 1

Peak Locate To Channel: 16384

Peak	Energy	Peak Size in	Peak CPS
No.	(keV)	Counts per Second	% Uncertainty
1	27.50	4.7375E+000	2.47
M 2	72.99	1.7887E+000	5.37
m 3	75.27	2.9677E+000	3.62
4	80.48	1.1429E+001	1.85
12	177.63	1.5761E+000	14.46
16	231.85	3.0540E+000	7.37
17	241.39	6.0479E+000	3.76
M 18	253.39	2.0465E-001	13.02
m 19	255.53	1.7536E-001	17.33
21	267.46	1.3538E-001	108.25
23	284.78	2.3365E+001	0.91
24	310.23	2.9799E-001	45.12

	27	343.42	3.8875E-001	32.56
	28	352.22	1.2069E-001	71.19
	30	373.10	7.3210E-003	1133.01
	31	384.50	2.1125E-001	35.56
	34	433.06	2.3596E-001	44.30
	35	452.89	2.9024E+000	4.36
M	36	460.05	1.7737E+000	3.45
m	37	463.51	9.0540E-001	5.63
M	40	487.75	4.9628E-001	9.58
m	41	492.80	1.8980E+001	0.61
	42	503.53	4.8694E-001	23.07
M	45	551.71	1.6316E-001	26.74
m	46	554.90	2.6635E-001	18.01
	47	586.81	7.8494E-001	13.71
	49	619.28	4.4294E-001	19.47
M	50	637.51	1.4129E+001	0.70
m	51	643.27	5.1686E-001	7.59
	52	665.55	1.5271E+000	5.70
	55	713.72	5.3030E-001	12.11
m	64	857.22	4.8784E-001	7.15
	65	910.66	1.0155E+000	7.31
	66	921.43	2.7016E-001	22.73
M	67	942.02	7.7390E-002	103.27
m	68	949.50	4.7650E-002	103.40
	69	1060.57	6.0924E-001	15.29
	70	1073.86	7.0419E-002	74.66

72	1149.69	1.2354E+000	8.31
74	1238.80	1.1673E-001	44.09
75	1267.91	6.7483E-002	63.73
76	1316.85	1.9686E-001	22.24
77	1370.29	8.8306E+001	0.26
78	1598.67	2.1400E-001	17.56
79	1648.47	2.3703E-001	16.29
80	1734.42	5.1247E+000	1.44
81	1891.14	1.6125E-001	24.57
82	2004.57	2.9856E-001	14.85
83	2218.63	2.9788E-002	109.61

Interference Corrected Activity Report 10/26/2011 8:55:05 AM

Peak No.	Energy (keV)	Channel	Peak Size in Counts per Second	Peak CPS % Uncertainty
84	2246.92		7.6488E+000	1.20
85	2310.17		8.5295E-003	385.04
86	2759.71		4.7780E+001	0.35

M = First peak in a multiplet region

m = Other peak in a multiplet region

F = Fitted singlet

Errors quoted at 1.000 sigma

Sample Genie-2000 Input File for a Common Research Reactor Taggant

***** G A M M A S P E C T R U M A N A L Y S I S

Filename: \\Nsc-server\home\user\CAMFILES\Dr. Charlton\tamu1.CNF

Report Generated On : 11/28/2012 4:42:21 PM

Sample Title : tamu1

Sample Description : CdTe 1 irradiated at tamu

Sample Identification : tamu1

Sample Type :

Sample Geometry : S36PS

Peak Locate Threshold : 3.00

Peak Locate Range (in channels) : 1 - 65535

Peak Area Range (in channels) : 1 - 65535

Identification Energy Tolerance : 3.000 keV

Sample Size : 1.000E+000 unit

Sample Taken On : 11/26/2012 3:52:00 PM

Acquisition Started : 11/26/2012 4:04:35 PM

Live Time : 3600.0 seconds
Real Time : 3651.9 seconds
Dead Time : 1.42 %

Energy Calibration Used Done On : 11/26/2012
Efficiency Calibration Used Done On : 6/19/2012
Efficiency ID : S1PS

Peak Analysis Report 11/28/2012 4:42:22 PM

Page 2

***** P E A K A N A L Y S I S R E P O R T *****

Detector Name: SH

Sample Title: tamu1

Peak Analysis Performed on: 11/28/2012 4:42:21 PM

Peak Analysis From Channel: 1

Peak Analysis To Channel: 8192

Peak	ROI	ROI	Peak	Energy	FWHM	Net Peak	Net Area
------	-----	-----	------	--------	------	----------	----------

	No.	start	end	centroid	(keV)	(keV)	Area	Uncert.
	1	136-	146	141.78	59.26	2.03	2.62E+004	419.08
	2	158-	182	177.23	74.30	1.47	5.07E+004	1192.32
	3	195-	211	200.51	84.18	1.52	1.55E+004	949.91
	4	215-	225	220.47	92.65	1.35	3.61E+005	875.83
	5	348-	360	354.43	149.50	1.97	3.54E+004	726.89
	6	371-	381	375.67	158.51	1.46	2.16E+004	619.77
	7	541-	553	547.25	231.32	1.41	1.43E+004	603.28
M	8	574-	596	580.10	245.26	1.37	2.29E+004	319.33
m	9	574-	596	593.07	250.77	1.38	7.58E+002	233.05
M	10	611-	661	616.75	260.81	1.33	3.35E+004	328.74
m	11	611-	661	630.92	266.83	1.34	1.06E+003	217.88
m	12	611-	661	646.12	273.28	1.34	3.23E+003	224.55
m	13	611-	661	657.46	278.09	1.35	7.89E+002	222.59
	14	740-	751	745.02	315.25	1.56	2.05E+003	474.15
	15	761-	771	767.81	324.92	0.85	1.48E+003	441.17
	16	788-	801	794.55	336.27	1.47	4.22E+005	828.80
	17	811-	820	813.90	344.48	1.44	2.11E+003	371.48
	18	826-	837	831.21	351.82	1.49	2.97E+003	418.97
	19	857-	867	860.95	364.44	1.32	1.85E+003	370.06
M	20	978-	1005	985.89	417.46	2.10	7.40E+003	220.06
m	21	978-	1005	999.00	423.02	2.11	1.40E+003	187.02
	22	1022-	1032	1025.80	434.40	1.44	7.20E+002	322.56
	23	1040-	1052	1045.85	442.90	1.71	3.57E+003	367.52
M	24	1063-	1092	1068.27	452.42	1.44	2.37E+003	160.85

m	25	1063-	1092	1085.05	459.54	1.44	1.11E+004	207.51
M	26	1143-	1178	1150.60	487.36	1.50	2.49E+003	157.61
m	27	1143-	1178	1162.33	492.33	1.51	9.06E+004	348.54
	28	1198-	1213	1206.39	511.03	2.76	7.90E+004	505.26
	29	1238-	1253	1246.01	527.85	1.52	2.94E+005	666.11
	30	1298-	1313	1305.60	553.13	2.29	7.73E+003	376.89
M	31	1430-	1472	1438.10	609.36	1.79	3.04E+003	154.67
m	32	1430-	1472	1456.26	617.07	1.80	1.27E+003	141.77
m	33	1430-	1472	1464.54	620.58	1.80	5.23E+003	169.83
	34	1711-	1724	1716.78	727.62	1.11	5.81E+002	299.21
	35	1826-	1848	1831.94	776.48	1.53	1.47E+003	449.96
	36	1871-	1884	1879.15	796.52	1.75	1.21E+003	295.05
	37	1922-	1937	1931.22	818.62	1.81	1.29E+003	324.54
	38	1947-	1961	1955.41	828.88	2.09	3.84E+003	314.43
	39	1993-	2006	1997.65	846.81	1.85	6.69E+003	304.56
	40	2458-	2468	2463.17	1044.35	1.15	6.54E+002	253.14
	41	2580-	2596	2587.87	1097.27	1.85	3.42E+003	357.58
	42	2636-	2651	2642.31	1120.37	2.47	1.12E+003	335.98

Peak Analysis Report

11/28/2012 4:42:22 PM

Peak No.	ROI start	ROI end	Peak centroid	Energy (keV)	FWHM (keV)	Net Peak Area	Net Area Uncert.
43	2659-	2678	2668.99	1131.69	3.19	8.30E+003	414.84
44	2692-	2710	2701.46	1145.47	2.05	2.05E+004	415.25

M	45	3040-	3076	3050.62	1293.64	2.03	1.72E+005	416.15
m	46	3040-	3076	3072.62	1302.97	2.04	4.94E+002	124.38
	47	3217-	3237	3227.44	1368.67	2.09	2.04E+005	599.77
	48	3437-	3454	3444.61	1460.83	2.15	5.31E+003	309.69
	49	3549-	3565	3555.00	1507.67	1.53	9.08E+002	262.14
	50	3862-	3883	3873.33	1642.76	2.28	4.08E+005	716.21
	51	3895-	3915	3905.47	1656.40	3.28	2.94E+004	327.28
	52	4073-	4094	4083.59	1731.99	2.33	1.26E+004	313.05
	53	4152-	4170	4160.67	1764.69	2.09	8.91E+002	268.59
	54	4261-	4273	4269.00	1810.66	2.17	1.03E+003	205.68
	55	4973-	4991	4980.46	2112.57	2.38	1.13E+003	148.73
	56	5098-	5121	5109.78	2167.45	2.57	4.31E+005	691.32
	57	5276-	5299	5287.53	2242.88	3.35	1.82E+004	205.28
	58	6155-	6175	6162.99	2614.39	2.77	1.49E+003	107.37
	59	6479-	6504	6491.75	2753.90	2.80	1.13E+005	342.02
	60	7262-	7281	7271.17	3084.65	0.92	1.39E+002	24.66

M = First peak in a multiplet region

m = Other peak in a multiplet region

F = Fitted singlet

Errors quoted at 1.000 sigma

Interference Corrected Activity Report 11/28/2012 4:42:23 PM

***** N U C L I D E I D E N T I F I C A T I O N R E P O R T

Sample Title: tamu1

Nuclide Library Used: H:\CAMFILES\Dr. Charlton\genie nuclide 1

..... IDENTIFIED NUCLIDES

Nuclide	Id	Energy	Yield	Activity	Activity
Name	Confidence	(keV)	(%)	(Bq /unit)	Uncertainty
NA-22	0.539	511.00*	181.00	1.06606E+004	6.30732E+002
		1274.55	99.90		
NA-24	1.000	1368.55*	100.00	1.34797E+005	4.32236E+003
		2754.05*	99.94	1.14255E+005	5.21694E+004
CL-38	0.999	1642.69*	31.00	1.39586E+007	1.19955E+006
		2167.68*	42.00	1.28325E+007	2.95282E+006
AR-41	1.000	1293.64*	99.16	4.68679E+005	1.41510E+004
CA-49	0.989	3084.40*	92.10	2.19119E+004	1.39864E+004
MN-56	1.000	846.75*	98.90	9.84431E+003	5.55676E+002
		1810.72*	27.20	8.80213E+003	2.01135E+003
		2113.05*	14.30	1.99932E+004	4.82078E+003
ZN-71	0.419	121.62	3.02		
		390.03	3.83		
		511.65*	30.20	5.36294E+008	8.70266E+007
		910.35	7.83		

		1120.07*	2.18	1.12479E+008	5.80434E+007
ZN-71m	0.332	386.38	93.00		
		487.36*	62.30	2.66470E+003	2.38483E+002
		511.65*	28.50	1.88367E+005	1.12940E+004
		620.29*	56.60	7.38095E+003	4.10891E+002
BR-82x	0.665	554.35*	70.80	3.30917E+003	2.41914E+002
		619.11*	43.40	3.97342E+003	2.24622E+002
		698.37	28.50		
		776.52*	83.50	6.81118E+002	2.09735E+002
		827.83*	24.00	6.46204E+003	5.81467E+002
		1044.00*	27.20	1.12921E+003	4.39052E+002
		1317.47	26.50		
		1474.88	16.32		
KR-85	0.751	151.18*	75.10	1.32876E+004	1.17265E+003
		304.86	13.70		
SR-85M	0.990	151.18*	12.40	3.12035E+005	2.76066E+004
		231.78*	84.10	2.31475E+004	2.35980E+003
CD-107	0.997	93.12*	4.70	1.65322E+006	6.94233E+004
		511.00*	0.40	9.49949E+006	5.62279E+005
		796.46*	0.06	1.20818E+006	3.10404E+005
		828.93*	0.16	1.63146E+006	1.44290E+005
CD-115	1.000	260.90*	1.94	2.84844E+005	2.61734E+004
		336.24*	45.90	1.82549E+005	1.59569E+004
		527.90*	27.40	2.99293E+005	1.80856E+004
IN-116	0.999	416.86*	28.90	6.85987E+004	5.83787E+003
		1097.30*	56.20	3.22982E+004	3.59789E+003

1293.54* 84.40 1.19687E+006 4.40351E+004

Interference Corrected Activity Report 11/28/2012 4:42:23 PM

Nuclide Name	Id Confidence	Energy (keV)	Yield (%)	Activity (Bq /unit)	Activity Uncertainty
SN-123M	0.943	160.32*	85.60	5.20368E+004	4.83113E+003
TE-123m	0.994	88.46	0.09		
		158.99*	84.00	2.94074E+003	2.73020E+002
TE-127m	0.692	57.63*	0.50	8.16261E+005	2.31891E+005
		88.26	0.08		
I-128	0.999	442.90*	16.90	1.55821E+005	2.48950E+004
TE-129	0.919	278.43*	0.57	2.06857E+005	6.12555E+004
		459.52*	7.70	3.13802E+005	2.18182E+004
		487.39*	1.42	3.98606E+005	3.55052E+004
		802.10	0.19		
		1083.85	0.49		
TE-131	0.920	149.72*	68.30	1.90932E+005	1.68694E+004
		452.33*	18.10	9.82319E+004	9.43401E+003
		492.67*	3.97	1.82280E+007	1.12915E+006
		602.05	4.13		
		997.24	3.27		
		1146.96*	4.84	6.05890E+006	2.28597E+005
ND-147	0.594	91.11*	27.90	1.44123E+005	8.29778E+003
		531.02	13.10		

DY-157	0.936	182.20	1.84		
		265.34*	0.19	1.48734E+005	5.14751E+004
		326.16*	92.00	4.73539E+002	1.54453E+002
DY-165	0.706	94.70*	3.60	4.92727E+006	5.85326E+005
		361.67*	0.84	1.90642E+005	9.95359E+004
		633.42	0.57		
		715.33	0.53		
		1079.62	0.09		
TM-170	1.000	84.30*	3.26	5.32807E+004	4.61234E+003

* = Energy line found in the spectrum.

@ = Energy line not used for Weighted Mean Activity

Energy Tolerance : 3.000 keV

Nuclide confidence index threshold = 0.30

Errors quoted at 1.000 sigma

Interference Corrected Activity Report 11/28/2012 4:42:23 PM

 ***** I N T E R F E R E N C E C O R R E C T E D R E P O R T *****

	Nuclide	Wt mean	Wt mean
Nuclide	Id	Activity	Activity

Name	Confidence	(Bq /unit)	Uncertainty
NA-22	0.539	7.106395E+003	1.279778E+003
NA-24	1.000	1.346566E+005	4.307605E+003
CL-38	0.999	1.379912E+007	1.111384E+006
AR-41	1.000	4.518097E+005	1.419008E+004
CA-49	0.989	2.191187E+004	1.398639E+004
MN-56	1.000	9.895058E+003	5.323357E+002
X FE-59	0.931		
X CU-64	0.985		
ZN-71	0.419	1.124791E+008	5.804343E+007
ZN-71m	0.332	1.480628E+003	2.392912E+002
BR-82x	0.665	2.197297E+003	1.283240E+002
KR-85	0.751	4.664580E+003	1.347075E+003
SR-85M	0.990	2.314746E+004	2.359796E+003
CD-107	0.997	1.100067E+006	1.335221E+005
CD-115	1.000	2.424993E+005	1.088219E+004
IN-116	0.999	4.307956E+004	2.960950E+003
? SN-123M	0.943	5.203683E+004	4.831130E+003
? TE-123m	0.994	2.940741E+003	2.730202E+002
X TE-127	0.795		
TE-127m	0.692	8.162611E+005	2.318907E+005
I-128	0.999	1.558209E+005	2.489498E+004
TE-129	0.919	2.705331E+005	1.992651E+004
X I-131	1.000		
TE-131	0.920	1.097418E+005	9.427864E+003

	ND-147	0.594	4.264613E+004	1.317082E+004
	DY-157	0.936	4.748740E+002	1.544524E+002
X	GD-159	0.986		
	DY-165	0.706	1.906415E+005	4.105698E+004
	TM-170	1.000	5.328073E+004	4.612342E+003
X	HG-203	0.978		

? = nuclide is part of an undetermined solution

X = nuclide rejected by the interference analysis

@ = nuclide contains energy lines not used in Weighted Mean Activity

Errors quoted at 1.000 sigma

Interference Corrected Activity Report 11/28/2012 4:42:23 PM

***** U N I D E N T I F I E D P E A K S *****

Peak Locate Performed on: 11/28/2012 4:42:21 PM

Peak Locate From Channel: 10

Peak Locate To Channel: 8192

Peak No.	Energy (keV)	Peak Size in Counts per Second	Peak CPS % Uncertainty	Peak Type
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	2	74.30	1.0122E+000	2.37	D-Esc.
M	8	245.26	4.6023E-001	1.40	Sum
m	9	250.77	1.5260E-002	30.74	Sum
m	12	273.28	6.4956E-002	6.96	D-Esc.
	14	315.25	3.0768E-002	31.08	Sum
	17	344.48	4.2404E-002	17.63	D-Esc.
	18	351.82	4.8977E-002	17.32	Sum
m	21	423.02	2.8212E-002	13.34	Sum
	22	434.40	1.4482E-002	44.83	Sum
M	31	609.36	4.6916E-002	6.73	S-Esc.
m	32	617.07	2.5546E-002	11.17	Sum
	34	727.62	1.0755E-002	56.21	Sum
	37	818.62	2.5926E-002	25.20	Sum
	43	1131.69	1.6704E-001	5.00	S-Esc.
m	46	1302.97	9.9395E-003	25.19	Sum
	48	1460.83	7.9903E-002	7.85	Sum
	49	1507.67	1.7294E-002	30.56	Sum
	51	1656.40	5.9184E-001	1.11	S-Esc.
	52	1731.99	2.5158E-001	2.51	D-Esc.
	53	1764.69	9.6310E-003	56.29	Sum
	57	2242.88	3.6578E-001	1.13	S-Esc.
	58	2614.39	1.5228E-002	14.50	

M = First peak in a multiplet region

m = Other peak in a multiplet region

F = Fitted singlet

Errors quoted at 1.000 sigma